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The overlooked pathway:

Hydraulic shortcuts and their influence on pesticide transport in agricultural areas

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Abstract

Introduction. Pesticides used in agriculture are transported to surface waters through various pathways and pose a major threat to aquatic ecosystems. Farmers and authorities take various measures to reduce pesticide transport to surface waters and to protect them from harmful effects. However, such actions can only be effective if the underlying processes driving the pesticide risk are understood well enough. Previous research suggests that so-called *hydraulic shortcuts* may be an important pesticide transport pathway that has been overlooked in the past. The term hydraulic shortcuts refers to inlet or maintenance shafts of agricultural storm drainage systems, but also to roads, farm tracks, channel drains, and ditches. Even though the relevance of hydraulic shortcuts for pesticide transport has been shown in single cases, it is unclear how often these structures occur and how relevant they are in general for pesticide transport compared to other pathways.

Objectives. This thesis aimed on quantifying the relevance of pesticide losses via hydraulic shortcuts to Swiss surface waters. For this, the following four research questions were investigated: 1) How often do hydraulic shortcuts occur in Swiss agricultural areas? 2) What is their relevance for surface runoff-related pesticide transport? 3) What is their relevance for spray drift-related pesticide transport? 4) What pesticide concentrations and loads are found in hydraulic shortcuts?

Occurrence of hydraulic shortcuts. To determine their occurrence, hydraulic shortcuts were systematically mapped in a set of Swiss arable land and vineyard catchments. The results show that hydraulic shortcuts are a frequent structure in Swiss arable land catchments and that inlet shafts are the most important shortcut type. Most of these inlet shafts belong to the storm drainage systems of roads and farm tracks and few of them are located directly in fields. With very few exceptions, all inlet shafts create a connectivity to surface waters via the underground pipe system, either directly (87%) or via wastewater treatment plants or combined sewer overflows (12%). In vineyards, the occurrence of shortcuts was found to be even higher than on arable land.

Relevance for surface runoff-related pesticide transport. To assess the relevance of shortcuts for surface runoff-related pesticide transport, surface runoff connectivity was modelled for twenty catchments representing arable land in Switzerland. The results show that in the analysed catchments for 43% to 74% of the agricultural areas with a surface runoff connectivity to surface waters, the connectivity is established by hydraulic shortcuts. An extrapolation to the national level shows similar results with 47% to 60% connected via hydraulic shortcuts. The results further imply that around half of the surface runoff from arable land and around half of the surface runoff-related pesticide load reaches surface waters via hydraulic shortcuts.

Relevance for spray drift-related pesticide transport. The relevance of shortcuts for spray drift-related pesticide transport was assessed for a set of arable land and vineyards catchments. For this, spray drift deposition to roads drained by shortcuts and to surface waters was modelled. The results show that for most analysed catchments, the drift to drained roads is much larger than the direct drift to surface waters, especially in vineyards. Compared to typical total pesticide loss fractions to surface waters, the spray drift losses to drained roads are rather small for arable land, but substantial in vineyards. Current literature suggests that during rain events major fractions of the drift deposited on roads can be washed off, especially for pesticides with low soil adsorption coefficients. Consequently, for such pesticides and particularly in vineyards, spray drift wash-off from drained roads is expected to be a major transport pathway to surface waters.

Measurements of concentrations and loads. Discharge and pesticide concentrations during rain events were measured in four out of 158 storm drainage inlets of a small Swiss catchment throughout a full agricultural season. These measurements were accompanied by additional measurements in the stream and by a collection of pesticide application data. The results show that agricultural storm drainage inlets strongly influence surface runoff and related pesticide transport in the studied catchment. High pesticide concentrations (up to 62 µg/L) were found in inlets and, during some rain events, transport through single inlets was responsible for up to 10% to the total stream load of certain pesticides. A rough extrapolation to the entire catchment suggests that during selected rain events, on average 30% to 70% of the load in the stream per pesticide was transported through inlets. Moreover, the results provide insights on the factors causing high pesticide transport through inlets.

Conclusions. Pesticide transport via hydraulic shortcuts is an important pathway for the pollution of Swiss surface waters that has been overlooked in the past. Current regulations and mitigation measures are not addressing this pathway and – consequently – fall short in preventing pesticide losses through this pathway. Pesticide transport via shortcuts should therefore be considered in the pesticide registration process and when designing regulations and mitigation measures. Moreover, the awareness of farmers on this transport process should be built and further research should focus on closing remaining knowledge gaps on hydraulic shortcuts.

Zusammenfassung

Einleitung. Pestizide aus der Landwirtschaft werden über verschiedene Eintragswege in Oberflächengewässer transportiert und beeinträchtigen die aquatischen Ökosysteme stark. Landwirte und Behörden ergreifen daher verschiedenste Massnahmen um den Pestizidtransport in Oberflächengewässer und das damit verbundene Risiko zu vermindern. Solche Massnahmen können jedoch nur dann effektiv sein, wenn die zugrundeliegenden Prozesse ausreichend verstanden sind. Die Forschung der vergangenen Jahre hat aufgezeigt, dass sogenannte *hydraulische Kurzschlüsse* einen wichtigen Eintragspfad für Pestizide in Oberflächengewässer darstellen können. Zu hydraulischen Kurzschlüssen gehören Einlauf- und Wartungsschächte von Regenentwässerungssystemen, aber auch Strassen, Wege, Einlaufrinnen und Entwässerungsgräben. Die Relevanz hydraulischer Kurzschlüsse für den Pestizidtransport in landwirtschaftlichen Einzugsgebieten wurde jedoch nur in Einzelfällen untersucht und es ist unklar wie wichtig sie generell im Vergleich zu anderen Eintragspfaden sind.

Ziele. Diese Arbeit hatte daher das Ziel, die Relevanz hydraulischer Kurzschlüsse für den Pestizidtransport in Schweizer Oberflächengewässer zu quantifizieren. Hierfür wurden die folgenden Forschungsfragen untersucht: 1) Wie häufig sind hydraulische Kurzschlüsse in Gebieten mit landwirtschaftlicher Nutzung in der Schweiz? 2) Wie relevant sind sie für den Pestizidtransport durch Abschwemmung? 3) Wie relevant sind sie für den Pestizidtransport durch Drift? 4) Welche Pestizid-Konzentrationen und -Frachten treten in hydraulischen Kurzschlüssen auf?

Häufigkeit hydraulischer Kurzschlüsse. Um die Häufigkeit hydraulischer Kurzschlüsse zu bestimmen, wurden diese für eine Reihe von Schweizer Ackerland- und Rebberg-Einzugsgebieten kartiert. Für Ackerlandgebiete zeigen die Resultate, dass hydraulische Kurzschlüsse häufig vorkommen und dass Einlaufschächte den wichtigsten Kurzschluss-Typ darstellen. Die meisten dieser Einlaufschächte gehören zu den Regenentwässerungssystemen von Strassen und Wegen. Einige wenige sind jedoch direkt auf den landwirtschaftlichen Flächen zu finden. Über das Leitungssystem der Regenentwässerung leiten mit wenigen Ausnahmen alle dieser Einlaufschächte in Oberflächengewässer ein – entweder direkt (87%) der über Kläranlagen und Mischwasserentlastungen (12%). In Rebberg-Einzugsgebieten kommen hydraulische Kurzschlüsse sogar noch häufiger vor.

Relevanz für Pestizidtransport durch Abschwemmung. Um die Relevanz von Kurzschlüssen für den Pestizidtransport durch Abschwemmung zu bestimmen, wurde die Oberflächenabfluss-Konnektivität in zwanzig Schweizer Ackerland-Einzugsgebieten modelliert. Die Resultate zeigen für die untersuchten Gebiete, dass von jenen landwirtschaftlichen Flächen, die eine Oberflächenabfluss-Konnektivität zu Oberflächengewässern haben, 43% bis 74% über hydraulische Kurzschlüsse ans Gewässer angeschlossen sind. Eine Extrapolation auf die nationale Ebene zeigt ähnliche Resultate mit 47% bis 60% der Flächen, die über Kurzschlüsse angeschlossen sind. Die Resultate deuten ausserdem darauf hin, dass rund die Hälfte des auf Ackerland gebildeten Oberflächenabflusses und des dadurch verursachten Pestizidtransportes über hydraulische Kurzschlüsse in die Gewässer gelangt.

Relevanz für Pestizidtransport durch Drift. Die Relevanz von Kurzschlüssen für den Pestizidtransport durch Drift wurde für eine Reihe von Ackerland- und Rebberg-Einzugsgebieten untersucht. Hierfür wurde die Driftdeposition auf durch Kurzschlüsse entwässerte Strassen und in Oberflächengewässer modelliert. Die Resultate zeigen, dass die Drift auf entwässerte Strassen in den meisten der untersuchten Gebiete und speziell in Rebbergen deutlich grösser ist als die direkte Drift in Oberflächengewässer. Im Vergleich zu typischen Gesamtverlustraten von Pestiziden ist die Driftdeposition auf entwässerten Strassen in Ackerlandgebieten eher klein, aber substanziell in Rebbergen. Aufgrund des aktuellen wissenschaftlichen Kenntnisstandes wird erwartet, dass während Regenereignissen ein grösserer Anteil der auf Strassen abgelagerten Drift abgewaschen wird, zumindest bei Pestiziden mit tiefen Bodenadorptionskoeffizienten. Für solche Pestizide und insbesondere in Rebbergen ist folglich damit zu rechnen, dass die Abwaschung der auf entwässerten Strassen abgelagerten Drift wesentlich zum Pestizidtransport in Oberflächengewässer beiträgt.

Messungen von Konzentrationen und Frachten. In einem kleinen landwirtschaftlichen Einzugsgebiet in der Schweiz wurden Abfluss und Pestizidkonzentrationen während Regenereignissen über eine volle landwirtschaftliche Saison hinweg gemessen. Die Messungen erfolgten in vier von insgesamt 158 Einlaufschächten im Gebiet und wurden durch weitere Messungen im Bach und durch Pestizid-Anwendungsdaten ergänzt. Die Resultate zeigen, dass die Einlaufschächte des landwirtschaftlichen Regenentwässerungssystems den Oberflächenabfluss im untersuchten Einzugsgebiet und den damit verbundenen Pestizidtransport stark beeinflussen. In den Einlaufschächten wurden hohe Pestizidkonzentrationen (bis zu 62 µg/L) gefunden. Einzelne Einlaufschächte waren während einigen Regenereignissen ausserdem für bis zu 10% der Gesamtfracht bestimmter Pestizide im Bach verantwortlich. Eine grobe Hochrechnung auf das gesamte Einzugsgebiet deutet darauf hin, dass während ausgewählten Regenereignissen durchschnittlich etwa 30% bis 70% der Pestizidfracht im Bach über Einlaufschächte transportiert wurde. Die Resultate zeigen ausserdem auf, welche Einflussfaktoren zu hohen Pestizidkonzentrationen in Einlaufschächten führen können.

Schlussfolgerungen. Der Transport durch hydraulische Kurzschlüsse ist ein wichtiger Eintragspfad für Pestizide in die Schweizer Oberflächengewässer. Die bestehenden gesetzlichen Bestimmungen und Reduktionsmassnahmen berücksichtigen diesen Transportprozess jedoch nicht und können Pestizideinträge über diesen Eintragspfad folglich nicht verhindern. Pestizideinträge über hydraulische Kurzschlüsse sollten daher im Pestizid-Zulassungsprozess berücksichtigt werden, sowie auch in den gesetzlichen Bestimmungen und bei Reduktionsmassnahmen. Darüber hinaus sollte bei Landwirten ein Bewusstsein für diesen Prozess geschaffen werden und zukünftige Forschung sollte darauf hinarbeiten, die verbleibenden Wissenslücken in Bezug auf hydraulische Kurzschlüsse zu schliessen.

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Chapter 1. Introduction

1.1 Pesticides – a global threat to aquatic ecosystems

Since decades, pesticides are used globally for the protection of agricultural crops. Together with increased irrigation and nitrogen and phosphorus fertilization, they have led to an increase in crop yields globally and are an important factor for global food production (Muller et al., 2017; Pingali, 2012; Tilman et al., 2001; Wauchope, 1978). However, pesticides applied in agriculture are also lost to non-target areas and organisms, posing a major threat to aquatic ecosystems and human health (Nicolopoulou-Stamati et al., 2016). They are one of the major contaminants of surface waters and groundwater, harm aquatic organisms, and cause biodiversity losses on the global scale (Malaj et al., 2014; Stehle and Schulz, 2015).

In Switzerland and other European countries, measures must be taken by law to protect surface waters against harmful effects, including the contamination by agricultural pesticides (EU Water Directive, 2000; GschG, 1992). Before the usage of a pesticide is permitted, it therefore has to undergo an extensive registration procedure. This procedure aims to ensure that permitted and correctly used pesticides are not posing an unacceptable risk for the environment, animals or humans. To further protect surface waters, farmers have to consider a broad range of regulations when applying pesticides. However, the registration procedure has increasingly been criticised for having fallen out of step with scientific knowledge and for not being able to sufficiently prevent the environment from adverse effects (Brühl and Zaller, 2019; Topping et al., 2020).

In accordance with this criticism, legal concentration limits and environmental quality standards are regularly exceeded in surface waters in Europe (Doppler et al., 2017; Halbach et al., 2021; Mohaupt et al., 2020), but also in groundwater used for drinking water production (Kiefer et al., 2020). To reduce the risk imposed by pesticides to the environment and to humans, many European countries have therefore formulated national action plans for risk reduction and sustainable use of pesticides (European parliament and Council of the European Union, 2009; WBF, 2017). The Swiss national action plan (WBF, 2017) – which was made legally binding by Parliament in 2021 – aims on a 50% reduction of the pesticide risks until 2027, and schedules a wide range of different actions to reach this goal. These actions include, inter alia, the promotion of reduced pesticide use (e.g. by alternative cultivation methods or plant varieties), the promotion of techniques and measures reducing pesticide transport to non-target areas (e.g. advanced cleaning systems for sprayers and additional guidelines for measures reducing runoff-related pesticide transport), and an increase of inspection, information, and consultation of farmers.

Actions aiming on a pesticide risk reduction can only be effective if the underlying processes driving the pesticide risk are understood well enough. However, recent field studies showed that there are still major knowledge gaps regarding how pesticides are transported from the field to surface waters. Especially, so-called hydraulic shortcuts were pointed out to potentially play an important role in this process (Doppler et al., 2012; Frey et al., 2009). These structures, however, have been largely overlooked in the past. Therefore, the Swiss national action plan also scheduled the execution of research projects to close specific knowledge gaps regarding pesticide risks, and (besides other research activities) this doctoral thesis was funded. Action 6.2.1.3 of the Swiss national action plan defined the scope of this doctoral thesis as follows: "[…] It is unclear which risks are imposed to surface waters by pesticide losses via hydraulic shortcuts (e.g. storm drainage systems of roads or farm tracks, or storm drainage inlet shafts on agricultural areas). These risks are also not considered in pesticide authorization. […] The project should therefore quantify the relevance of pesticide losses to surface waters via hydraulic shortcuts. Additionally, measures for the reduction of pesticide losses via hydraulic shortcuts should be identified." (WBF, 2017)

This doctoral thesis aims on answering the above-mentioned question raised by the authorities. The following sections give an introduction on the current state of research related to pesticide transport processes to surface waters (Sect. 1.2.1) and provide a definition of the term *hydraulic shortcuts* (Sect. 1.2.2). Afterwards, current research gaps on pesticide transport via hydraulic shortcuts are emphasized (Sect. 1.3), and the objectives and an outline of this thesis are described (Sect. 1.4).

1.2 Pesticide loss pathways to surface waters

1.2.1 Classical pathways

In current literature, the pathways causing pesticide losses to surface waters (see Figure 1) are usually divided into point sources and diffuse sources (Holvoet et al., 2007; Reichenberger et al., 2007). Point sources are related to urban water infrastructure, i.e. *wastewater treatment plants* (1) (Munz et al., 2017) and *sewer overflows* (2) (Mutzner et al., 2020; Wittmer et al., 2010), but also include losses related to *bad management practices* (3) (Reichenberger et al., 2007), such as farmyard runoff after the filling or washing of spraying equipment. As the most important diffuse sources, the following pathways are considered: *Surface runoff* (4) is formed on fields during rain events and can transport pesticides in dissolved form, or sorbed to eroded soil particles (Holvoet et al., 2007; Larsbo et al., 2016; Lefrancq et al., 2017). By *preferential flow through soil macropores* (5), pesticides can reach tile drainages and quickly be transported to surface waters (Accinelli et al., 2002; Sandin et al., 2018). In the case of *spray drift* (6), a part of the droplets originating from the spraying process, can be transported with the wind to surface waters (Carlsen et al., 2006; Stephenson et al., 2006; Vischetti et al., 2008). Finally, also *atmospheric deposition* of volatilized pesticides or *aeolian deposition* (i.e. wind deposition of eroded soil particles) can be important diffuse transport pathways under certain conditions and for specific environments (Bish et al., 2021; Jones et al., 2019). In contrast, leaching to groundwater and subsequent exfiltration to surface waters is usually considered negligible.

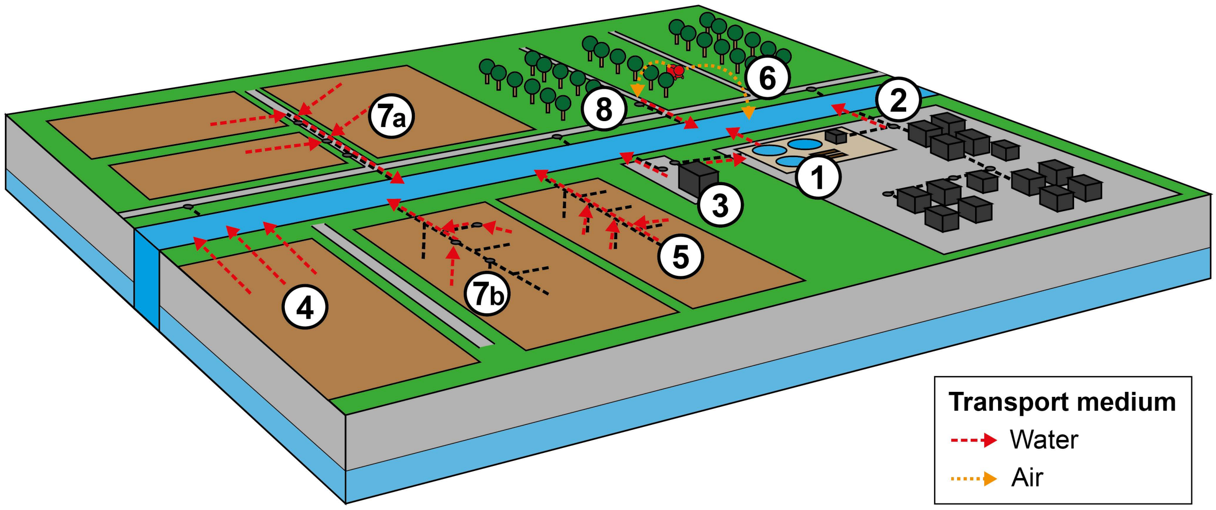

Figure 1: Most important classical pesticide loss pathways to surface waters (1-6), and different pathways related to hydraulic shortcuts (7-8). Wastewater treatment plants (1), sewer overflow (2), losses due to bad management, e.g. farmyard runoff (3), direct surface runoff (4), preferential flow to tile drainages (5), direct spray drift (6), indirect surface runoff – e.g. via road storm drainage inlets (7a) or storm drainage inlets in the field (7b), indirect spray drift (8).

1.2.2 Hydraulic shortcuts

Linear landscape structures, such as roads, ditches, or hedges have an important influence on pesticide transport in catchments (Fiener et al., 2011; Payraudeau et al., 2009). Hedges have been shown to intercept surface runoff and spray drift, and therefore to reduce pesticide transport to surface waters. In contrast, roads and (roadside) ditches have been shown to promote concentrated runoff and runoff connectivity in catchments, and therefore to increase the surface runoff and pesticide transport to the stream (Carluer and De Marsily, 2004; Hösl et al., 2012). At the same time, roads and ditches may also collect spray drift from nearby applications (Meli et al., 2007). Studies in a French and a German vineyard catchment showed that the wash-off of spray drift deposited on roads led to high pesticide concentrations in road runoff during the rain events following pesticide application. Additionally, this process was reported to be responsible for a large fraction of the runoff-related pesticide load at the catchment outlet (Lefrancq et al., 2014; Rübel, 1999).

Especially in Switzerland, many roads and farm tracks in agricultural areas are drained by inlet shafts of road storm drainage systems (Alder et al., 2015). Such inlet shafts are sometimes even located directly in fields. These inlet shafts, but also maintenance shafts of the storm water and tile drainage systems have been shown to further increase the surface runoff and pesticide connectivity in catchments (Doppler et al., 2012).

Structures enhancing the transport of surface runoff and agricultural pollutants to surface waters have been referred to as *hydraulic shortcuts* or *short-circuits* (Carluer and De Marsily, 2004; Doppler et al., 2014; Frey et al., 2009). Within this thesis, a hydraulic shortcut is defined as follows:

> A *hydraulic shortcut* is an artificial structure that increases and/or accelerates the process of surface runoff reaching surface waters (i.e. rivers, streams, lakes) or makes this process possible in the first place.

Hydraulic shortcuts therefore include the following structures: a) roads and farm tracks, b) storm drainage inlet shafts, c) maintenance shafts, and d) channel drains and ditches. However, if such a structure is present in the landscape, this is referred to as a *potential* shortcut. Only if this potential shortcut is effectively creating connectivity to surface waters, it is considered a *real* shortcut.

Sometimes also tile drainage systems are considered hydraulic shortcuts (e.g. Gassmann et al. (2013)). However, surface runoff and pesticides need to pass through a soil layer to reach the tile drainage system. Also, pesticide transport linked to tile drainages has been investigated in many studies and is also recognised as a relevant transport pathway in the pesticide registration process. Therefore, this transport pathway was not considered as a shortcut within this thesis.

Based on the preceding elaborations, pesticide losses to surface waters via hydraulic shortcuts can be summarized into two categories. These categories are illustrated in Figure 2 and are explained in the following (numbers relate to the processes shown in Figure 1). In the case of *indirect surface runoff* (7), surface runoff is formed on crop areas, flows to a shortcut structure, and is then directed to surface waters. In contrast, surface runoff that is directly flowing to surface waters, is referred to as direct surface runoff in this thesis. In the case of *indirect spray drift* (8), spray drift is deposited on roads, farm tracks, or other hard surfaces during application and is then washed-off to surface waters during the next rain event. The washed-off surface runoff can either reach the stream by flowing along roads, farm tracks, or other hard surfaces, or by being transported through a storm water drainage system. In contrast, spray drift that is transported directly by the wind to surface waters, is referred to as direct spray drift in this thesis.

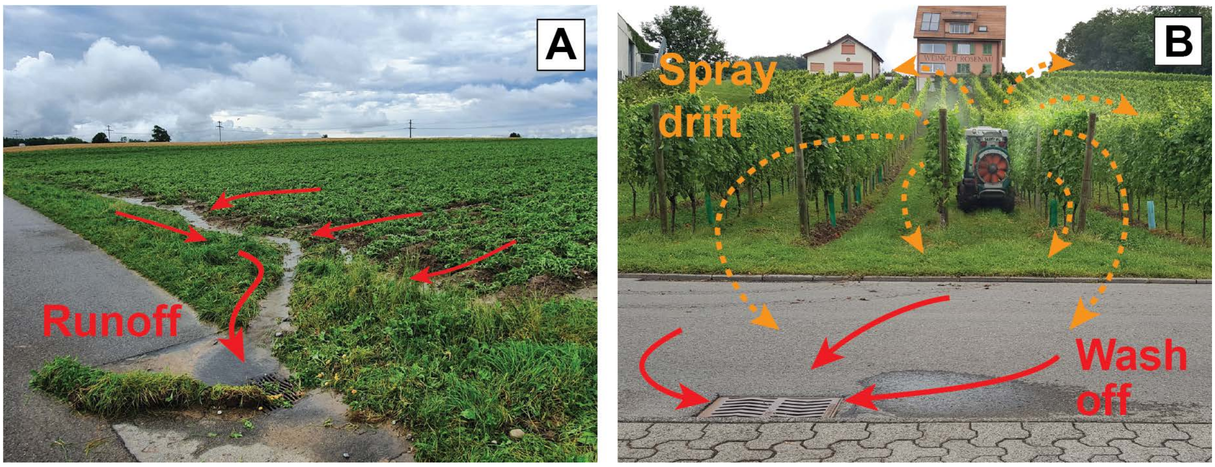

Figure 2: A – Illustration of indirect surface runoff on the example of a field in the canton of Zürich, shortly after a heavy rain event. B – Illustration of indirect spray drift on the example of a vineyard in the canton of Lucerne.

1.3 Research gaps

The preceding elaborations show that hydraulic shortcuts may largely influence the pesticide transport in agricultural catchments. To answer the questions raised by the authorities regarding the relevance of hydraulic shortcuts on the national scale, various research gaps have to be closed. These gaps are explained in the following.

1) **How often do hydraulic shortcuts occur in Switzerland?**

   The spatial occurrence of roads and farm tracks in Switzerland is well known due to the availability of a national spatial dataset in high quality (Swisstopo, 2020a). However, the occurrence of other shortcuts (inlet shafts, maintenance shafts, channel drains, ditches) has only been systematically assessed and reported in one study. For a an area of 3.6 km^2 in western Switzerland, Prasuhn and Grünig (2001) mapped 268 storm drainage inlet shafts on arable land, corresponding to a density of around 0.8 inlet shafts per hectare of agricultural land. Due to the small area analysed and since the study was only restricted to one region, it remains unknown if the findings of this study can be generalized. Consequently, there is a need of a systematic and representative assessment of the occurrence of hydraulic shortcuts at the national scale.

2) **What is the relevance of indirect surface runoff for pesticide transport?**

   To assess the relevance of indirect surface runoff for pesticide transport, in the study previously mentioned (Prasuhn and Grünig, 2001), also the influence of hydraulic shortcuts on surface runoff connectivity was analysed. For the study region, 62% of the agricultural area was estimated to be connected indirectly to surface waters, and 3.2% directly. Similarly, for a small agricultural catchment (1.2 km^2) in north-eastern Switzerland, Doppler et al. (2012) reported 23% of the agricultural area to be connected indirectly to surface waters, whereas only 4.4% was connected directly. While these studies were only restricted to small areas, two further studies showed similar results for larger scales using a modelling approach. For the canton of Basel-Landschaft, Bug and Mosimann (2011) reported 35% of the agricultural area to be connected indirectly, and 12.5% directly to surface waters. On the national scale, Alder et al. (2015) reported 34% to be connected indirectly and 21% directly. However, in these studies, the occurrence of hydraulic shortcuts was not explicitly known and was modelled based on generalizing assumptions (e.g. classification of roads as either drained or undrained, based on their size). These assumptions however underlie major uncertainties and it is unclear how they influence the surface runoff connectivity estimates. Therefore, there is a need for a surface runoff connectivity assessment on the national scale, based on spatially explicit, representative data on the occurrence of hydraulic shortcuts.

3) What is the relevance of indirect spray drift for pesticide transport?

Only few studies have assessed the influence of indirect spray drift on pesticide transport to surface waters. Rübel (1999) reported for a German vineyard catchment that wash-off from vineyard roads after helicopter applications led to high pesticide concentrations in the receiving stream. Similarly, Lefrancq et al. (2014) reported for air blast sprayer applications in a French vineyard catchment that spray drift on roads and subsequent wash-off was a major pathway for fungicide transport to the catchment outlet. For a Swiss catchment with a predominance of arable crops, Ammann et al. (2020) showed – based on the field study of Doppler et al. (2012) – that the uncertainty of exposure models could be strongly reduced by considering spray drift wash-off from roads. However, the spatial structure (e.g. density of roads around crop areas) and the types and number of shortcuts present are expected to be different in Swiss catchments compared to the catchments analysed in Rübel (1999) and Lefrancq et al. (2014). Moreover, the amount of spray drift deposited on roads depends largely on the spraying method used (e.g. Rautmann et al. (1999)), and accordingly on the predominant crop types in a catchment. Therefore, the potential of indirect spray drift should be assessed for a representative set of catchments and for different crop types.

4) What pesticide concentrations and loads are found in hydraulic shortcuts?

Field measurements on the transport of agricultural pollutants through inlet and maintenance shafts have only been performed in two studies. Firstly, in a long-term study focusing on soil erosion, Remund et al. (2021) showed that 88% of the phosphorus and sediment losses to surface waters occurred via inlet or maintenance shafts. Secondly, Doppler et al. (2012) showed that these structures allowed for a fast transport of pesticides between remote areas in the catchment and the stream. However, direct measurements of surface runoff, pesticide concentrations, or pesticide loads in inlet or maintenance shafts of agricultural storm drainage systems do not exist up to now. To fill this gap, pesticide transport through inlet and maintenance shafts should be measured.

1.4 Objectives and thesis content

The goal of this thesis was to quantify the relevance of pesticide losses via hydraulic shortcuts to Swiss surface waters compared to other transport pathways. Additionally, the thesis aimed on proposing measures for the reduction of pesticide losses via hydraulic shortcuts.

The remaining part of this thesis is divided into four chapters. Chapters 2 to 4 focus on closing the research gaps pointed out in Sect. 1.3 as outlined in the following. Since these chapters are all published in scientific journals, they needed to be readable independently. Therefore, some repetitions may occur, especially in the introduction sections. Finally, chapter 5 presents an overall conclusion on the research presented in this thesis.

Chapter 2: Occurrence of hydraulic shortcuts and indirect surface runoff

In this chapter, the occurrence of hydraulic shortcuts was assessed by a mapping campaign in twenty arable land catchments in the Swiss midlands. Additionally, their influence on surface runoff connectivity and related pesticide transport were quantified using a modelling approach.

Chapter 3: Indirect spray drift

Similar to chapter 2, the research in this chapter was based on a combination of field mapping with a modelling approach. Pesticide transport by indirect spray drift was modelled in 26 catchments (17 with a predominance of arable cropping, and 9 with vineyards). For this, data from the field mapping campaign of chapter 2 was used and an additional campaign for vineyards was performed.

Chapter 4: Field measurements on pesticide transport in storm drainage inlets

In contrast to the previous two chapters, actual measurements on pesticide transport were performed for this chapter. In a small catchment in the Swiss midlands with intensive agricultural use, surface runoff, pesticide concentrations, and pesticide loads were measured in inlets of the storm water drainage system and compared to the same measurements in the receiving stream. These measurements were performed for one agricultural season and were combined with plot-specific application data to identify important processes and other factors influencing pesticide transport through inlets.

Chapter 2. Hydraulic shortcuts increase the connectivity of arable land areas to surface waters

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Abstract

Surface runoff represents a major pathway for pesticide transport from agricultural areas to surface waters. The influence of artificial structures (e.g. roads, hedges, and ditches) on surface runoff connectivity has been shown in various studies. In Switzerland, so-called hydraulic shortcuts (e.g. inlet and maintenance shafts of road or field storm drainage systems) have been shown to influence surface runoff connectivity and related pesticide transport. Their occurrence and their influence on surface runoff and pesticide connectivity have, however, not been studied systematically.

To address that deficit, we randomly selected 20 study areas (average size of 3.5 km^2) throughout the Swiss plateau, representing arable cropping systems. We assessed shortcut occurrence in these study areas using three mapping methods, namely field mapping, drainage plans, and high-resolution aerial images. Surface runoff connectivity in the study areas was analysed using a 2×2 m digital elevation model and a multiple-flow algorithm. Parameter uncertainty affecting this analysis was addressed by a Monte Carlo simulation. With our approach, agricultural areas were divided into areas that are either directly, indirectly (i.e. via hydraulic shortcuts), or not at all connected to surface waters. Finally, the results of this connectivity analysis were scaled up to the national level, using a regression model based on topographic descriptors, and were then compared to an existing national connectivity model.

Inlet shafts of the road storm drainage system were identified as the main shortcuts. On average, we found 0.84 inlet shafts and a total of 2.0 shafts per hectare of agricultural land. In the study catchments, between 43 % and 74 % of the agricultural area is connected to surface waters via hydraulic shortcuts. On the national level, this fraction is similar and lies between 47 % and 60 %. Considering our empirical observations led to shifts in estimated fractions of connected areas compared to the previous connectivity model. The differences were most pronounced in flat areas of river valleys.

These numbers suggest that transport through hydraulic shortcuts is an important pesticide flow path in a landscape where many engineered structures exist to drain excess water from fields and roads. However, this transport process is currently not considered in Swiss pesticide legislation and authorization. Therefore, current regulations may fall short in addressing the full extent of the pesticide problem. However, independent measurements of water flow and pesticide transport to quantify the contribution of shortcuts and validating the model results are lacking. Overall, the findings highlight the relevance of better understanding the connectivity between fields and receiving waters and the underlying factors and physical structures in the landscape.

2.1 Introduction

Agriculture has been shown to be a major source for pesticide contamination of surface waters (Loague et al., 1998; Stehle and Schulz, 2015). Pesticides are known to pose a risk to aquatic organisms and to cause biodiversity losses in aquatic ecosystems (Beketov et al., 2013; Malaj et al., 2014). For implementing effective measures to protect surface waters from pesticide contamination, the relevant transport processes have to be understood.

Pesticides are lost to surface waters through various pathways from either point sources or diffuse sources. In current research, surface runoff (Holvoet et al., 2007; Larsbo et al., 2016; Lefrancq et al., 2017), preferential flow through macropores into the tile drainage system (Accinelli et al., 2002; Leu et al., 2004a; Reichenberger et al., 2007; Sandin et al., 2018), and spray drift (Carlsen et al., 2006; Schulz, 2001; Vischetti et al., 2008) are considered of major importance. Other diffuse pathways like leaching into groundwater and exfiltration into surface waters, atmospheric deposition or aeolian deposition are usually less important.

Past research showed that different catchment parts can largely differ in their contribution to the overall pollution of surface waters (Gomides Freitas et al., 2008; Leu et al., 2004b; Pionke et al., 1995). This is the case for soil erosion or phosphorus, but also for pesticides. Areas largely contributing to the overall pollution load are called critical source areas (CSAs). Models delineating such CSAs assume that those areas fulfill three conditions (Doppler et al., 2012): i) They represent a substance source (e.g. pesticides, soil, phosphorus), ii) they are connected to surface waters, and iii) they are hydrologically active (e.g. formation of surface runoff).

Linear landscape structures, such as hedges, ditches, tile drains, or roads have been shown to be important features for the connectivity within a catchment (Fiener et al., 2011; Rübel, 1999). Undrained roads were reported to intercept flow paths, to concentrate and accelerate runoff, and therefore also to influence pesticide connectivity within a catchment (Carluer and De Marsily, 2004; Dehotin et al., 2015; Heathwaite et al., 2005; Payraudeau et al., 2009). Additionally, Lefrancq et al. (2013) showed that undrained roads act as interceptor of spray drift, possibly leading to significant pesticide transport during subsequent rainfall events when intercepted pesticides are washed off the roads.

However, such linear structures and the related connectivity effects exhibit substantial regional differences due to natural conditions or various aspects of the farming systems. In contrast to other countries, many roads in agricultural areas in Switzerland are drained by stormwater drainage systems (Alder et al., 2015). Inlet shafts of stormwater drainage systems are also found directly in fields (Doppler et al., 2012; Prasuhn and Grünig, 2001). Since those stormwater drainage systems were reported to shortcut surface runoff to surface waters, those structures were called *hydraulic shortcuts* or short-circuits. Doppler et al. (2012) showed in a small Swiss agricultural catchment that hydraulic shortcuts were creating connectivity of remote areas to surface waters and had a strong influence on pesticide transport. Only 4.4% of the catchment area was connected directly to surface waters, while 23% was connected indirectly (i.e. via hydraulic shortcuts). For the same catchment, Ammann et al. (2020) showed that the uncertainty of a pesticide transport model could be reduced by 30% by including catchment-specific knowledge about hydraulic shortcuts and tile drainages.

The occurrence of hydraulic shortcuts and their influence on catchment connectivity has only been studied for a few other catchments in Switzerland. Prasuhn and Grünig (2001) found that only 3.2% of the arable land in five small catchments were connected directly to surface waters, while 62% were connected indirectly. Consequently, 90% of the sediment lost to surface waters was transported through shortcuts.

To our knowledge, these two studies are the only ones systematically assessing the occurrence of hydraulic shortcuts and their influence on (sediment) connectivity. However, since these studies only covered a small total area in specific regions, it remains unknown if these findings are generally valid for Swiss agricultural areas.

Two other studies in Switzerland addressed connectivity on a larger scale using a modelling approach. Both indicated that more areas were connected through shortcuts than directly. Bug and Mosimann (2011) estimated 12.5% of the arable land in the canton of Basel-Landschaft to be connected directly to surface waters, and 35% to be connected indirectly. Later, Alder et al. (2015) created a national connectivity map of erosion risk areas. They estimated that 21% of the agricultural area is connected directly to surface waters and 34% indirectly. Since only for small areas the occurrence of hydraulic shortcuts was effectively known, generalizing assumptions on the occurrence of hydraulic shortcuts were made in both studies (e.g. classification of roads as drained by shortcuts or as undrained, based on their size). As also stated by Alder et al. (2015), these assumptions are a major source of uncertainty. Their influence on the estimated connectivity fractions remains unclear.

In summary, previous studies on hydraulic shortcuts were either restricted to small study areas in a specific region, or were based on generalizing assumptions, lacking a spatially explicit consideration of hydraulic shortcuts. This study aims for a systematic, spatially distributed, and representative assessment of hydraulic shortcut occurrence on Swiss agricultural areas. Based on this assessment we aim on quantifying the influence of hydraulic shortcuts on surface runoff connectivity and pesticide transport. Additionally, we aim on estimating how additional data on the occurrence of shortcuts influence the connectivity fractions reported by the existing national connectivity map. We focused our study on arable land, since this is the largest type of agricultural land with common pesticide application in Switzerland.

Our research questions therefore are:

1) How widespread do hydraulic shortcuts occur in Swiss arable land areas?
2) What is the contribution of hydraulic shortcuts to surface runoff connectivity and what are potential implications for surface-runoff related pesticide transport?

3) How are additional data on the occurrence of shortcuts influencing the connectivity predictions at the national scale?

2.2 Material and Methods

2.2.1 Selection of study areas

We selected 20 study areas (Table 1) representing arable land in the Swiss plateau and the Jura mountains (Figure 3). This selection was performed randomly on a nationwide small-scale topographical catchment dataset (BAFU, 2012). The probability of selection was proportional to the total area of arable land in the catchment as defined by the Swiss land use statistics (BFS, 2014). Random selection was performed using the pseudo-random number generator Mersenne Twister (Matsumoto and Nishimura, 1998).

On average, the study areas have a size of 3.5 km^2 and are covered by 59% agricultural land. The agricultural land mainly consists of arable land (74%) and meadows/pastures (21%). The mean slope on agricultural land is 4.9 degrees and the mean annual precipitation amounts to 1159 mm yr^{-1}. A comparison of important catchment properties of the study areas to the corresponding distribution of all Swiss catchments with arable land demonstrated that the study areas represent the national conditions well (see Figure S1).

Table 1: Catchment properties of the 20 study areas. Fractions of agricultural area and of arable land were determined from BFS (2014). Mean slope of agricultural areas was determined from BFS (2014) and Swisstopo (2018). Mean annual precipitation was determined from Kirchhofer and Sevruk (1992).

ID	Location	Can-ton	Receiving water	Area (km^2)	Fraction of agri-cultural area	Fraction of arable land	Mean slope of agri-cultural areas (deg)	Mean an-nual preci-pitation (mm/yr)
1	Böttstein	AG	Bruggbach	3.3	52%	30%	8.5	1187
2	Ueken	AG	Staffeleggbach	2.0	42%	39%	7.6	1164
3	Rüti b. R.	BE	Biberze	2.2	29%	11%	11.2	1403
4	Romont	FR	Glaney	3.4	78%	48%	4.0	1344
5	Meyrin	GE	Nant d'Avril	10.0	49%	31%	3.2	1133
6	Boncourt	JU	Saivu	5.9	44%	23%	5.5	1093
7	Courroux	JU	Canal de Bellevie	2.8	82%	75%	2.9	1082
8	Hochdorf	LU	Stägbach	2.4	84%	59%	4.1	1213
9	Müswangen	LU	Dorfbach	3.0	79%	61%	4.0	1482
10	Fleurier	NE	Buttes	1.0	24%	11%	9.6	1538
11	Lommiswil	SO	Bellacher Weiher	3.8	50%	40%	6.8	1388
12	Illighausen	TG	Tobelbach	1.9	54%	30%	1.8	1122
13	Oberneunforn	TG	Brüelbach	3.3	69%	52%	4.2	968
14	Clarmont	VD	Morges	2.4	75%	70%	5.3	1163
15	Molondin	VD	Flonzel	4.2	74%	65%	5.9	1064
16	Suchy	VD	Ruis.des Combes	3.3	72%	63%	5.6	1026
17	Vufflens	VD	Venoge	2.8	39%	30%	5.7	1006
18	Buchs	ZH	Furtbach	3.9	57%	48%	4.9	1182
19	Nürensdorf	ZH	Altbach	2.3	59%	44%	3.6	1225
20	Truttikon	ZH	Niederwisenbach	5.1	66%	49%	4.6	960
			Mean	**3.5**	**59%**	**44%**	**4.9**	**1159**

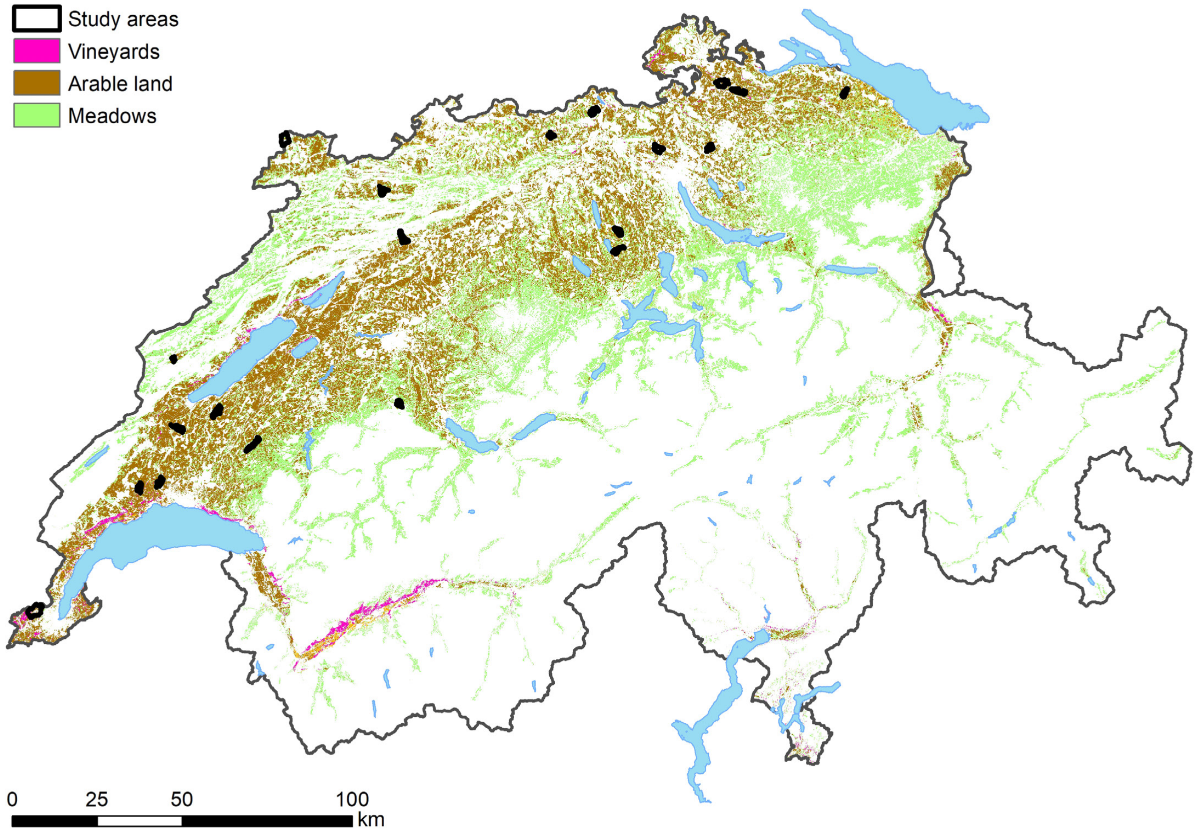


Figure 3: Study areas (black) and distribution of arable land (brown), vineyards (pink), and meadows/pastures (green) across Switzerland. Source: BFS (2014); Swisstopo (2010)

2.2.2 Assessment of hydraulic shortcuts

Shortcut definition

We define a hydraulic shortcut as *an artificial structure increasing and/or accelerating the process of surface runoff reaching surface waters (i.e. rivers, streams, lakes) or making this process possible in the first place*. In this study, we focused on the following structures (example photos can be found in Figure S2 to Figure S13):

A) Storm drainage inlet shafts on roads, farm tracks and crop areas
B) Maintenance shafts of storm drainage systems or tile drainage system on roads, farm tracks and crop areas
C) Channel drains and ditches on roads, farm tracks and crop areas

If one of these structures is present, we defined this as a *potential shortcut*. If surface runoff can enter the structure and if the structure is drained to surface waters or to a wastewater treatment plant, this is defined as a *real shortcut*. Other processes that are sometimes referred to as hydraulic shortcuts (e.g. tile drains) are not considered in this study. Tile drains have already received considerable attention in pesticide research and the transport to tile drains includes flow through natural soil structures.

Shortcut location and type

We mapped the location and types of potential shortcuts in each study area by combining three different methods.

i) *Field survey:* Field surveys were performed between August 2017 and May 2018 (details see Table S5). In a subpart of each study area, we walked along roads and paths and mapped all the potential shortcut structures. The starting point was selected randomly, and we mapped as much as we could within one day. Consequently, the field survey data only cover a part of the catchment. For each of the potential shortcuts we recorded its location, as well as a set of properties using a smartphone and the app "Google My Maps". This included a specification of the type of the shortcut (e.g. inlet, inspection chamber, ditches, channel drains), its lid type (e.g. grid, sealed lid, lid with small openings), and its lid height relative to the ground surface. A list of all possible types can be found in the supporting information (Table S2 to Table S4).

ii) *Drainage plans:* For all municipalities covering more than 5% of a study area we asked the responsible authorities to provide us with their plans of the road storm drainage systems and the agricultural drainage systems. For 38 and 26 of the 46 municipalities concerned we received road storm drainage system plans and tile drainage system plans, respectively. Reasons for missing data are either that the responsible authorities did not respond or that data on the drainage systems were not available. From the plans, we extracted the locations of shortcuts and, if available, the same properties were specified as in the field survey.

iii) *Aerial images:* Between August 2017 and August 2018 we acquired aerial images of the study areas with a ground resolution of 2.5 to 5 cm (details see Table S5). We used a fixed-wing UAV (eBee, Sensefly, Cheseaux-sur-Lausanne) in combination with a visible light camera (Sony DSC-WX220, RGB). The study areas were fully covered by the UAV imagery, with the exception of larger settlement areas, forests, and lakes, and of no-fly zones for drones (e.g. airports). The UAV images were processed to one georeferenced aerial image per study area using the software Pix4Dmapper 4.2. In the no-fly zones of the study areas Meyrin (Geneva), Buchs (Zürich), and Nürensdorf (Zürich) we used aerial images provided by the cantons of Geneva (Etat de Genève, 2016) and Zürich (Kanton Zürich, 2015). Ground resolutions were 5 cm, and 10 cm respectively. Using ArcGIS 10.7, we gridded the aerial images, scanned by eye through each of the grid cells, and marked all potential shortcut structures manually. If observable from the aerial image, the same properties as for the field survey were specified for each potential shortcut structure.

We combined the three datasets originating from the three methods to a single dataset. If a potential shortcut structure was only found by one of the mapping methods, its location and type were used for the combined dataset. If it was found by more than one of the mapping methods, we used the location and type of the mapping method that we expected to be the most accurate. For the location information, this is UAV imagery, before field survey, and maps. For the type specification, this is field survey, before UAV imagery, and maps.

Assigning shortcuts to different landscape elements

In order to better understand where hydraulic shortcuts occur the most, we assigned them to different landscape elements. Using the topographic landscape model of Switzerland “swissTLM3D” (Swisstopo, 2010) we defined five landscape elements: Paved roads, unpaved roads, fields, settlements, and other areas (e.g. railways, other traffic areas, forests, water bodies, wetlands, single buildings). For all landscape elements except roads and railways, shortcuts were assigned to their landscape elements by a simple intersection. However, shortcuts belonging to road or railway drainage systems are in many cases not placed on the road or railway directly, but on the adjacent agricultural land or settlement. Therefore, shortcuts were assigned to the landscape elements road or railway if they were within a 5 m buffer.

In addition, we correlated the density of shortcuts per study area to different study area properties. We selected study area properties that we expected to have explanatory power: density (length per area) of paved roads, density of unpaved roads, density of surface rivers, density of subsurface rivers, mean annual precipitation, and mean slope on agricultural areas.

Drainage of shortcuts

A potential shortcut only turns into a real one if it is drained to surface waters by pipes or other connecting structures, such as ditches. Therefore, using the plans provided by the municipalities, we investigated where potential shortcuts drain to. They were allocated to one of the following categories of recipient areas: surface waters, wastewater treatment plants/combined sewer overflow, infiltration areas (e.g. forest, infiltration ponds, fields, grassland), or unknown.

2.2.3 Surface runoff connectivity model

To assess how hydraulic shortcuts contribute to surface runoff connectivity, we created a surface runoff connectivity model.

The model is based on the concept of critical source areas (CSAs, see introduction). It mainly focuses on the first two elements of the CSA concept (pesticide application and connectivity to surface waters). In contrast, the question whether an area is hydrologically active is only addressed partially because many relevant information such as soil properties are not available at the national scale.

The model (see Figure 4) distinguishes *source areas* on which surface runoff is produced, and *recipient areas* on which surface runoff ends up. A *connectivity model* connects those areas by routing surface runoff through the landscape. These model parts are conceptually described in more detail in the section “model structure”. In the section “model parametrization”, we describe how we parametrized the model and how we assessed the uncertainty of model output given the parameter uncertainty. In the last section "hydrological activity", we explain the testing for systematic differences in the hydrological activity between areas with direct or indirect connectivity.

Model structure

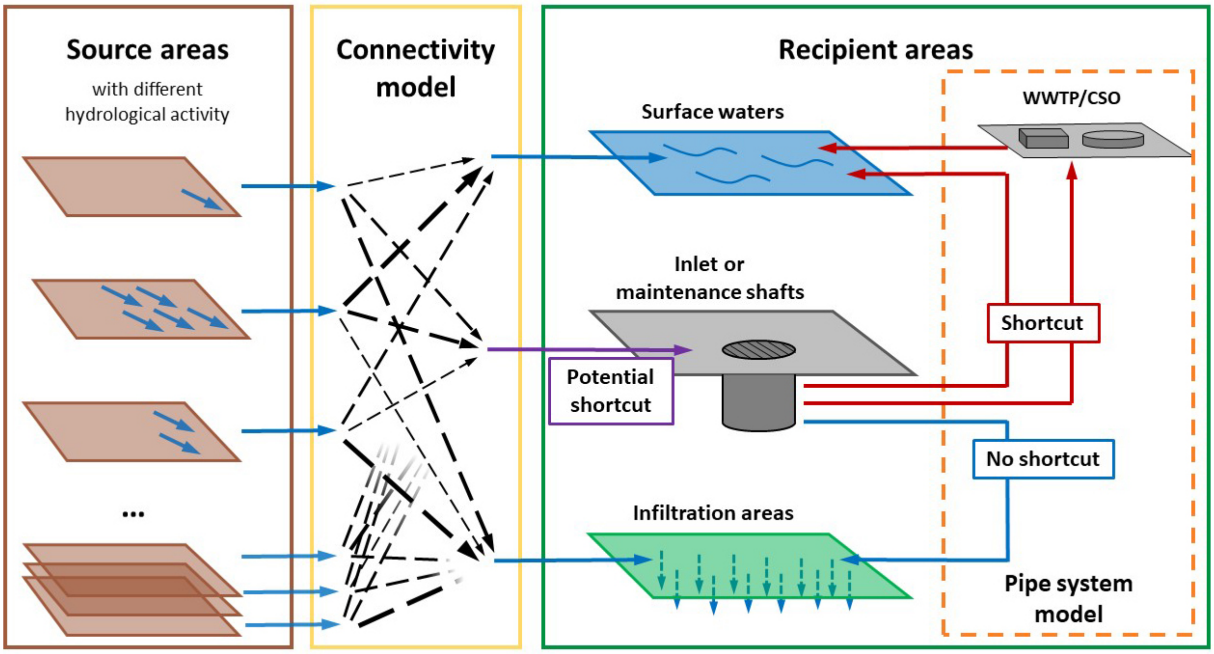


Figure 4: Structure of the surface runoff connectivity model. WWTP: Waste water treatment plants, CSO: Combined sewer overflow.

Source areas. All crop areas on which pesticides are applied should in theory be considered as source areas. However, a highly resolved spatial dataset of land in a crop rotation for our study areas is lacking. Therefore, we considered the total extent of agricultural areas (i.e. arable land, meadows/pastures, vineyards, orchards, and gardening) as source areas, since those areas could be derived in high resolution. The extent of agricultural areas was defined by subtracting all non-agricultural areas from the extent of the study area. For this, we used non-agricultural areas (forests, water bodies, urban areas, traffic areas, and other non-agricultural areas) as defined by the national topographical landscape model SwissTLM3D (Swisstopo, 2010). According to the Swiss proof of ecological performance (PEP), pesticide usage within a distance of 6 m from a river, and within 3 m from hedges and forests is prohibited. The extent of agricultural areas was reduced accordingly except along forests (parameters *river spray buffer*, *hedge spray buffer*).

Recipient areas. Surface runoff generated on a source area and routed through the landscape can end up in three different types of landscape elements, referred to as recipient areas: Surface waters, infiltration areas (i.e. forests, hedges, internal sinks), and shortcuts. The extent of surface waters (rivers that have their course above the surface, lakes, and wetlands), was defined by the SwissTLM3D model as was the extent of forests and hedges. Since forests and hedges are known to infiltrate surface runoff (Bunzel et al., 2014; Dosskey et al., 2005; Schultz et al., 2004; Sweeney and Newbold, 2014) we assumed that forests with a certain width (parameter *infiltration width*) act as an infiltration area. Hedges were assumed either to act as infiltrations areas, or to have no effect on surface runoff. Accordingly, the parameter *hedge infiltration*, was varied between yes (hedges act as infiltration areas) and no (hedges don't act as an infiltration areas).

Internal sinks in the landscape were defined using the 2 x 2 m digital elevation model (Swisstopo, 2018). All sinks larger than two raster cells and deeper than a certain depth (parameter *sink depth*) were defined as internal sinks. All other sinks were filled completely.

Shortcuts were defined in two different ways (parameter *shortcut definition*): In definition A, all inlet shafts, ditches, and channel drains were considered as potential shortcuts. In definition B, maintenance shafts lying in internal sinks were additionally considered as potential shortcuts. Potential shortcuts were defined to act as real shortcuts if they are known to discharge to surface waters or wastewater treatment plants. From the drainage plans of the municipalities, we know that most of the inlet shafts discharge into either a surface water body or a wastewater treatment plant. Therefore, also potential shortcuts with unknown drainage location were assumed to act as real shortcuts. Potential shortcuts discharging into forests or infiltration structures were assumed not to act as shortcuts and were not used in the model. Shortcut recipient areas were defined as the raster cells of the digital elevation model on which the shortcut is located and all the cells directly surrounding it (see Figure S14).

Connectivity model. For modelling connectivity we used the TauDEM model (Tarboton, 1997) which is based on a D-infinity flow direction approach. As an input we used a 2 x 2 m digital elevation model (DEM) (Swisstopo, 2018). This DEM was modified as follows: We assumed that only those internal sinks that were defined as sink recipient areas (see above) effectively act as sinks. Therefore, firstly, all sinks were filled, and sink recipient areas were carved 10 m into the DEM. Secondly, all other recipient areas (shortcuts, forests, hedges, surface waters) were carved between 10 and 50 m into the DEM. Carving the recipient areas into the DEM ensured that surface runoff reaching a recipient area was not routed further on to another recipient area. Thirdly, to account for the effect of roads accumulating surface runoff (Heathwaite et al., 2005), roads were carved into the DEM by a given depth defined by the parameter *road carving depth*.

The modified DEM, the source areas, and the recipient areas were used as an input into the TauDEM tool "D-Infinity upslope dependence". Like this, each raster cell belonging to a source area was assigned with a probability to be drained into one of the three types of recipient areas.

The connectivity of a source area may depend on the flow distance to surface waters. For longer flow distances, water has a higher probability to infiltrate before it reaches a surface water. Therefore, for each source area raster cell, we calculated the flow distance to its recipient area using the tool "D-infinity distance down".

Model parametrization and sensitivity analyses

The model parameters mentioned in the section above vary in space and time. Since this variability could not be addressed with the selection of a single parameter value, we performed a Monte Carlo simulation with 100 realizations. The probability distributions of the parameters are provided in Table 2. The bounds or categories of these distributions were based on our prior knowledge about the hydrological processes involved, about structural aspects (e.g. depths of sinks), and on our experience from field mapping. The parameters *river spray buffer* and *hedge spray buffer* were assumed constant according to the guidelines of the Swiss proof of ecological performance (PEP).

To assess the influence of single parameters on our modelling results, we performed a local sensitivity analysis against a benchmark model (one realization of the model with a specific parameter set, see Table 2). When selecting the benchmark model parameter set, we kept the changes in the digital elevation model small (i.e. *road carving depth* = 0 cm, *sink depth* = 10 cm). For the other model parameters, we selected the values that we assumed to be the most probable in reality. For the local sensitivity analysis, each of the model parameters was varied individually within the same boundaries as for the Monte Carlo analysis.

Table 2: Summary of parameter distributions used for the Monte Carlo analysis and parameter values used as a benchmark for the sensitivity analysis. PEP: Swiss proof of ecological performance.

Parameter	Handling of parameter uncertainty	Distribution	Bounds / Categories	Benchmark model
Sink depth	Monte Carlo & sensitivity analysis	Uniform distribution	5 cm ≤ x ≤ 100 cm	10 cm
Infiltration width	Monte Carlo & sensitivity analysis	Uniform distribution	6 m ≤ x ≤ 100 m	20 m
Road carving depth	Monte Carlo & sensitivity analysis	Uniform distribution	0 cm ≤ x ≤ 100 cm	0 cm
Shortcut definition	Monte Carlo & sensitivity analysis	Bernoulli distribution	[Definition A; Definition B]	Definition A
Hedge infiltration	Monte Carlo & sensitivity analysis	Bernoulli distribution	[yes; no]	Yes
River spray buffer	Assumed as certain, based PEP guidelines	Constant	6 m	6 m
Hedge spray buffer	Assumed as certain, based PEP guidelines	Constant	3 m	3 m

Hydrological activity

As mentioned earlier, a critical source area has to be hydrologically active, i.e. surface runoff has to be generated on that area. Runoff generation depends on many variables (e.g. crop types, soil types, soil moisture, rain intensity) for which no data are available in most of our study areas and which are strongly variable over time. Since we are interested in the general relevance of shortcuts, we focused on the question whether there is a systematic difference in the hydrological activity between areas directly or indirectly connected to streams.

For soil moisture, we tested for such differences by calculating the distribution of the topographic wetness index (TWI) (Beven and Kirkby, 1979) for the source areas of the benchmark model. We calculated the TWI as follows, using the "Topographic Wetness Index" tool of the TauDEM model:

$$\mathrm{TWI} = \frac{\ln(a)}{\tan(\beta)} \tag{2.1}$$

The local upslope area a, and the local slope β were calculated using the D-infinity flow direction algorithm that was already used for the surface runoff connectivity model. As an input, we used the source areas and the modified DEM as specified for the surface runoff connectivity model.

The formation of surface runoff on agricultural areas is also influenced by their slope. Therefore, we calculated the distribution of slopes for source areas draining to different destinations. For this we used the slopes from the Swiss digital elevation model (Swisstopo, 2018).

For other variables (e.g. crop type, rain intensity), there is no indication for such systematic differences. Therefore, we assumed that they do not differ systematically between areas draining to different recipient areas.

2.2.4 Extrapolation to the national level

Extrapolation of the local connectivity model

In a last step, we developed a model for extrapolating the results from our study areas (local surface runoff connectivity model, LSCM) to the national scale. This extrapolation was then used to evaluate how the results of this study compare to a pre-existing connectivity model (Alder et al., 2015).

Selection of explanatory variables: We calculated a list of catchment statistics based on nationally available geodatasets that could serve as explanatory variables. As catchment boundaries, the polygons from the national catchment dataset (BAFU, 2012) were used. Details on the datasets used for calculating those catchment statistics can be found in Table S1.

We created a linear regression between each of those catchment statistics to the median fractions of agricultural areas directly, indirectly, and not connected to surface waters, as reported by the LSCM ($f_{LSCM,dir}$, $f_{LSCM,indir}$, $f_{LSCM,nc}$). The strongest correlations were found for the fractions of agricultural areas directly, indirectly, and not connected to surface waters, as reported by the NECM ($f_{NECM,dir}$, $f_{NECM,indir}$, $f_{NECM,nc}$, see Table S8). Therefore, we used them as explanatory variables for building an extrapolation model of our local results to the national scale.

The model predictions for each catchment have to fulfil specific boundary conditions: Firstly, the sum of areal fractions of the three types of recipient areas k per catchment c has to equal one ($\sum_{k=1}^{K} f_{k,c} = 1$), and secondly, area fractions cannot be negative ($f_{k,c} \geq 0$). To ensure these conditions, we performed the model fit after a unit simplex data transformation. To address the uncertainty introduced by the selection of our study catchments, we additionally bootstrapped the model one hundred times. The resulting modelling approach is shown in Figure 5. Mathematical details are provided in Sect S2.1.5.

As a result, we obtained a national surface runoff connectivity model (NSCM). The NSCM provides an estimate for the fractions of agricultural areas directly, indirectly, and not connected to surface waters ($f_{NSCM,dir}$, $f_{NSCM,indir}$, $f_{NSCM,nc}$) for the catchments of the national catchment dataset. Since in the NECM mountainous regions of higher altitudes are excluded, those areas are also excluded in the NSCM.

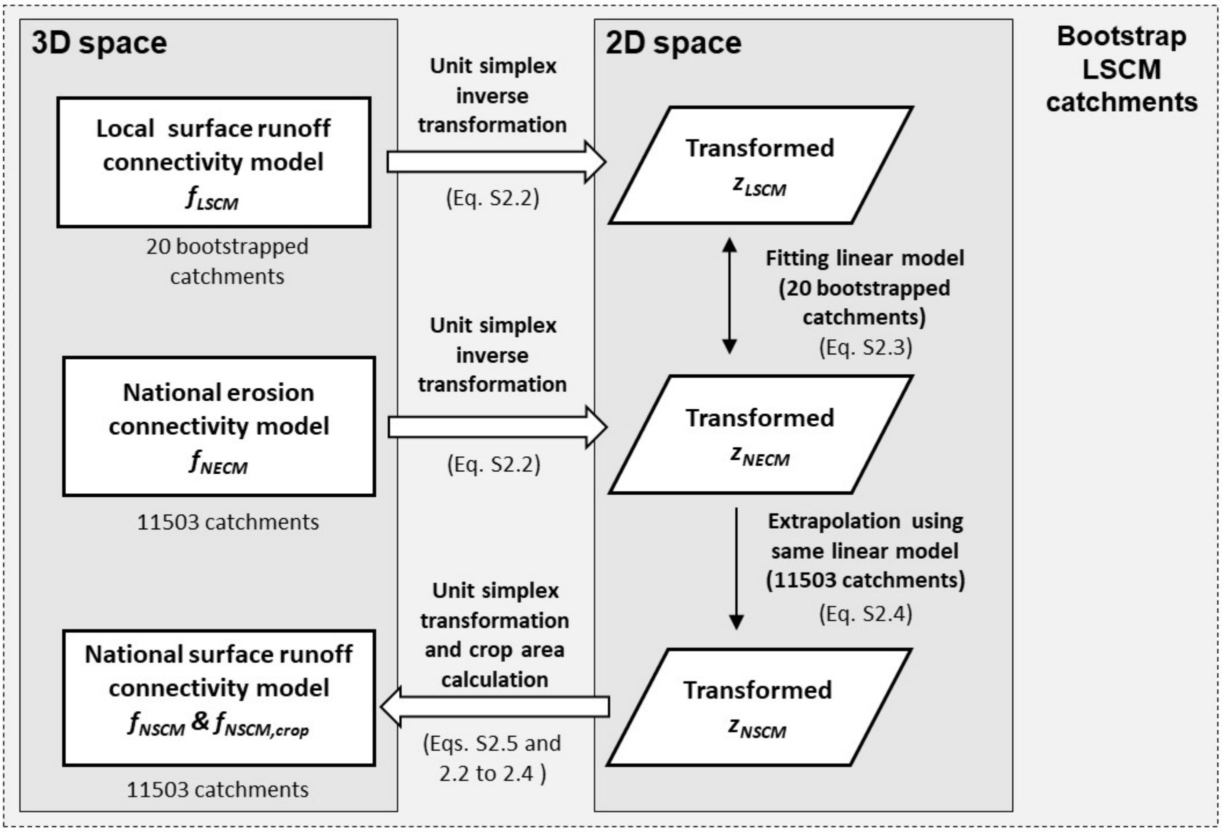


Figure 5: Extrapolation of the local surface runoff connectivity model (LSCM) to the national scale (NSCM) using a unit simplex transformation approach.

Connectivity of crop areas

During the time of this study, high-resolution datasets of Swiss crop areas were not available in Switzerland. Therefore, we considered the total extent of agricultural areas for building the local surface runoff connectivity model and extrapolation to the national scale. This includes areas with rare pesticide application, such as meadows and pastures.

The Swiss land use statistics dataset (BFS, 2014) is a raster dataset with a resolution of 100 m, dividing agricultural areas into different categories (e.g. arable land, vineyards, meadows/pastures). On the national scale, the usage of such a lower-resolution dataset is more reasonable. Hence, we used this dataset for calculating fractions of connected crop areas.

The fractions of directly, indirectly, and not connected crop areas per total agricultural area per catchment c ($f_{NSCM,crop,c}$) were calculated as follows:

$$f_{NSCM,crop,c} = f_{NSCM,c} \cdot r_{crop,c} \tag{2.2}$$

With r_{crop} being the ratio of crop area to total agricultural area in a catchment:

$$r_{crop,c} = \frac{A_{crop,c}}{A_{crop,c}+A_{mead,c}} \tag{2.3}$$

$$A_{crop,c} = A_{arab,c} + A_{vin,c} + A_{orch,c} + A_{gard,c} \tag{2.4}$$

with: $A_{crop,c}$ = Crop area in catchment c (ha)

$A_{mead,c}$ = Meadow and pasture areas in catchment c (ha)

$A_{arab,c}$ = Arable land area in catchment c (ha)

$A_{vin,c}$ = Vineyard area in catchment c (ha)

$A_{orch,c}$ = Orchard area in catchment c (ha)

$A_{gard,c}$ = Gardening area in catchment c (ha)

2.3 Results

2.3.1 Occurrence of hydraulic shortcuts

In the following section, we first show the results of the field mapping campaign for shafts (inlet shafts, maintenance shafts) followed by the results for channel drains and ditches. Afterwards we present results on the accuracy of our mapping methods.

Shafts

In total, we found 8213 shafts, corresponding to an average shaft density of 2.0 ha^{-1} (min.: 0.51 ha^{-1}, max.: 4.4 ha^{-1}; Table 3). Forty-two percent of the shafts mapped were inlet shafts. A plot showing the density of shafts mapped per catchment and shaft type can be found in Figure S15 in the supporting information.

For roughly half of the inlet and maintenance shafts we were able to identify a drainage location. Both shaft types discharge in almost all cases into surface waters, either directly (87% of inlet shafts, 63% of maintenance shafts) or via wastewater treatment plants or combined sewer overflow (12% of inlet shafts, 37% of maintenance shafts). Only 1.4% of the inlet shafts and no maintenance shaft at all, were found to drain to an infiltration area, such as forests or fields.

Most of the inlet shafts mapped (90%) are located on paved or unpaved roads (see Table 4). Only very few inlet shafts (2.8%) are found directly on fields. In contrast, maintenance shafts are found much more often on fields and therefore less often on paved or unpaved roads. The fractions of inlet shafts and maintenance shafts belonging to a certain landscape element for each study area can be found in Figure S18 in the supporting information.

We correlated the densities of inlet and maintenance shafts per study area with possible explanatory variables. Only the density of paved roads was significantly correlated to the density of inlet shafts ($R^2 = 0.33$, $p = 0.008$) and maintenance shafts ($R^2 = 0.37$, $p = 0.005$) (see Table S6 and Table S7).

Table 3: Number of shafts found on agricultural areas of the study areas per shortcut category and drainage location.

Drainage location	Inlet shafts		Maintenance shafts		Other shafts		Unknown type	
	Count	Fraction	Count	Fraction	Count	Fraction	Count	Fraction
Surface waters	1568	46%	1205	29%	0	0%	0	0%
WWTP/CSO	218	6%	705	17%	0	0%	0	0%
Infiltration areas	26	1%	0	0%	0	0%	0	0%
Unknown	1615	47%	2227	54%	31	100%	618	100%
Total	3427	100%	4137	100%	31	100%	618	100%

Table 4: Percentage of shafts found on a certain type of landscape element. The category "other areas" integrates several types of landscape elements: railways, other traffic areas, forests, water bodies, wetlands, and single buildings.

Shaft type	Paved roads	Unpaved roads	Settlements	Fields	Other areas
Inlet shafts	79%	10%	5.5%	2.8%	2.2%
Maintenance shafts	52%	7.2%	16%	21%	4.5%

Channel drains and ditches

In addition to shafts, we also mapped channel drains and ditches. With the exception of the study areas Meyrin (4.2 m ha^{-1}) and Buchs (4.0 m ha^{-1}) these structures were rarely found (< 1.2 m ha^{-1}; see Figure S16). In Meyrin and Buchs, most channel drains and ditches (98% of the total length) drain to surface waters, and only few of them to infiltration areas (2%).

Mapping accuracy

The results above were generated using three different mapping methods (*field survey*, *UAV images*, and *drainage plans*). These methods differ in their ability to identify and classify a potential shortcut structure correctly and in the study area they cover. We determined the accuracy of the mapping methods aerial images and drainage plans using the field survey method as a ground truth (see Table 5) for those parts of the study areas where all three methods were applied. Since channel drains and ditches were rare, this assessment was only performed for shafts.

The recall (i.e. the probability that a potential shortcut is found by a mapping method) was limited for the aerial images method (53% for inlet shafts, and 62% for maintenance shafts), and even lower for the drainage plans method (32% for inlet shafts, and 21% for maintenance shafts). However, identified shortcuts were in most of the cases classified correctly (accuracy: 93% to 94% for aerial images, 88% to 89% for drainage plans).

For the entire study areas, Figure 6 shows the number of potential shortcuts identified by the three mapping methods. Despite a low recall, aerial images identified the largest number of potential shortcuts. This is due to the large spatial coverage by the aerial images method. Since the overlap between the three methods is small (only 32% of the inlet shafts and 15% of the maintenance shafts were found by more than one method), each of the methods was important to determine the total number of potential shortcuts in the study areas. Because the aerial images and drainage plans have a low recall, but cover large parts of the study areas that were not assessed by the field survey, the numbers reported above are a lower boundary estimate.

Table 5: Recall and classification accuracies of the mapping methods aerial images and drainage plans. The recall corresponds to the probability that a potential shortcut is found by the mapping method. Percentages indicate the recall of each individual mapping method. In brackets, the recall of the combination of both methods is given. The accuracy corresponds to the sum of true positive fraction and true negative fraction.

Mapping method	Shaft type	Identification	Classification				
		Recall	True positives	False positives	True negatives	False negatives	Accuracy
Aerial images	Inlet	53% (60%)	61%	1.3%	33%	4.9%	94%
	Maintenance	62% (69%)	32%	5.3%	61%	1.3%	93%
Drainage plans	Inlet	32% (60%)	67%	4.5%	22%	6.6%	89%
	Maintenance	21% (69%)	20%	7.1%	68%	5.3%	88%

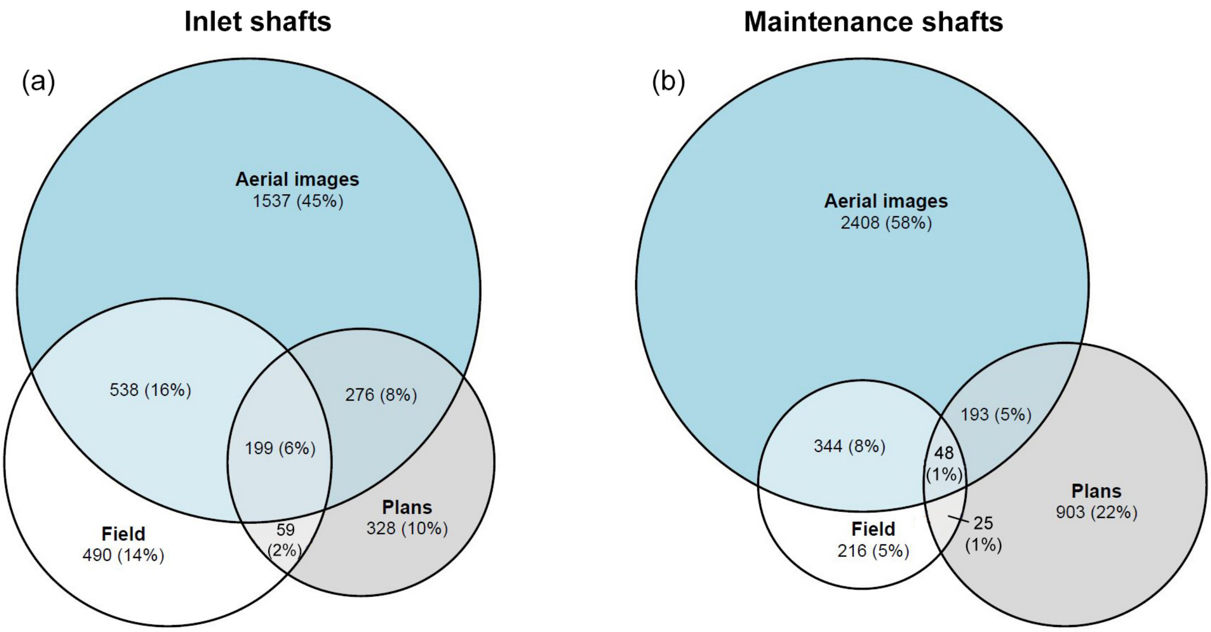
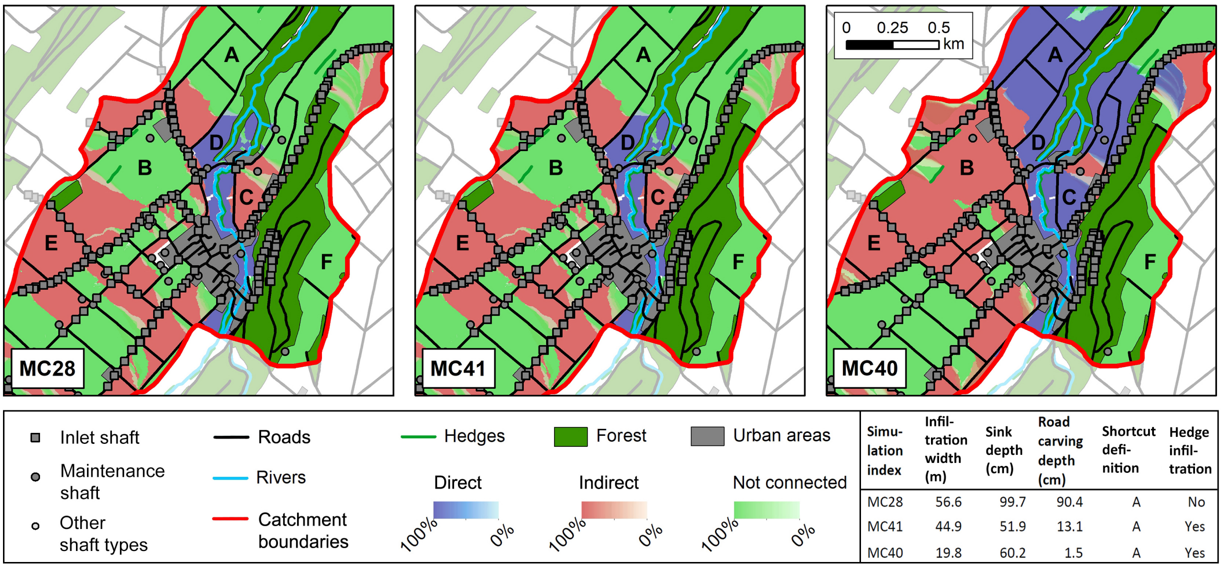


Figure 6: Number of inlet shafts (left) and maintenance shafts (right) identified by the different mapping methods.

2.3.2 Surface runoff connectivity

2.3.2.1 Study areas

From the Monte Carlo analysis of the surface runoff connectivity model, we obtained an estimate for the fractions of agricultural areas that are connected directly, indirectly, or not at all to surface waters. To illustrate the variability resulting from these Monte Carlo (MC) runs, Figure 7 shows the output of three MC simulations (MC28, MC41, and MC40) for Molondin. These simulations correspond to the 5%, 50%, and 95% quantile of the median fraction of indirectly connected per total connected agricultural area over all study catchments. The classification of certain catchment parts is changing depending on the model parametrisation (e.g. letters A to C). However, for other parts, the results are consistent across the different MC simulations (e.g. letters D to F). Overall, the results show that not only agricultural areas close to surface waters (e.g. letter D) are connected to surface waters. Hydraulic shortcuts also create surface runoff connectivity for areas far away from surface waters (e.g. letter E).



Simulation index	Infiltration width (m)	Sink depth (cm)	Road carving depth (cm)	Shortcut definition	Hedge infiltration
MC28	56.6	99.7	90.4	A	No
MC41	44.9	51.9	13.1	A	Yes
MC40	19.8	60.2	1.5	A	Yes

Figure 7: Results of three example Monte Carlo (MC) simulations for a part of the study area Molondin. The color ramps show the probability of agricultural areas to be directly connected (blue), indirectly connected (red) and not connected (green). The simulations represent approximately the 5% (MC28), 50% (MC41), and 95% (MC40) quantiles with respect to the resulting median fractions of indirectly connected per total connected area over all study catchments. The parameters of the example MC simulations are shown on the bottom right. Source of background map: Swisstopo (2010)

In order to assess the importance of hydraulic shortcuts, we calculated the fraction of indirectly connected area to the total connected area. Across all Monte Carlo simulations, the median of this fraction over all study catchments ranges between 43% and 74% (mean: 57%, median: 58%; Figure 8). Despite considerable uncertainty, the results demonstrate that a large fraction of the surface runoff connectivity to surface waters is established by hydraulic shortcuts.

For different flow distances, the fraction of indirectly connected area to the total connected area underlies only minor variations (see Figure S24). However, this fraction varies strongly between the study areas, with median fractions ranging from 21% in Müswangen to 97% in Boncourt. Although the occurrence of hydraulic shortcuts is a prerequisite of indirect connectivity, high shaft densities are not necessarily leading to high fractions of indirect connectivity in a catchment. The densities of inlet shafts and maintenance shafts show only a weak positive correlation to the catchment medians of the fraction of indirectly connected areas (inlet shafts: $R^2 = 0.11$, $p = 0.15$; maintenance shafts: $R^2 = 0.08$, $p = 0.23$; see Table S8). By contrast, the two study areas with high channel drain and ditch densities (Meyrin and Buchs) show high fractions of indirect connectivity. Similarly, the density of surface waters is strongly negatively correlated to the fraction of indirect connectivity ($R^2 = 0.51$, $p < 0.001$). This suggests that line elements like channel drains, ditches and surface waters usually have an influence on connectivity if they occur in a catchment. By contrast, the influence of point elements seems to depend a lot on the surrounding landscape structure.

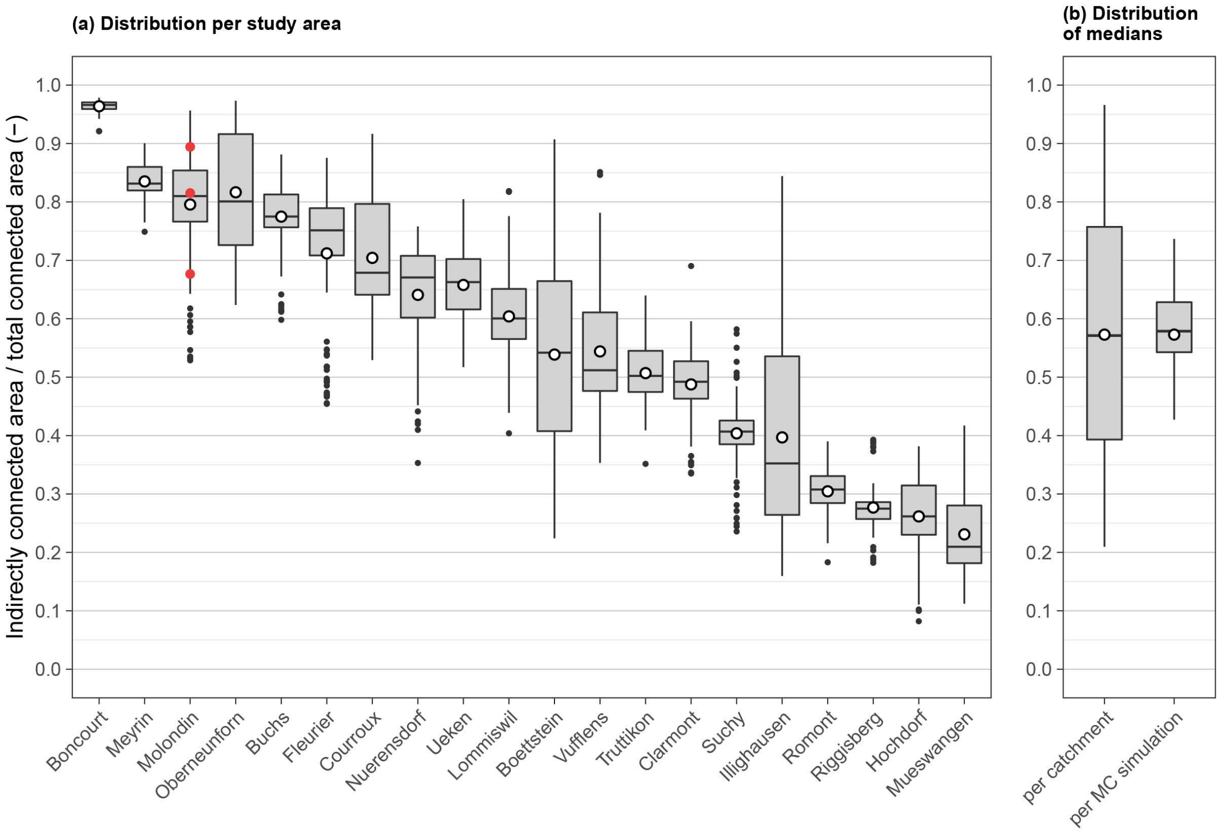


Figure 8: (a): Fractions of indirectly connected areas per total connected areas as calculated by the Monte Carlo analysis for each study area. White dots indicate the means of the distributions. The red dots indicate the results of the example Monte Carlo simulations (MC28, MC41, and MC 40) shown in Figure 7. (b): Distribution of medians of fractions of indirectly connected areas per total connected areas per study catchment and per Monte Carlo simulation.

As a further consequence of the structural differences between the study areas, not all of them reacted the same way to changes in model parameters of the Monte Carlo analysis. For example, the fraction of indirectly to total connected areas in the study area Boncourt was quite insensitive to changes in model parameters. Since Boncourt has a very low water body density, only small areas are connected directly, independent of the model parametrization. The study area Illighausen, on the other hand, reacted very sensitively (range of results = 68%). Since Illighausen is a very flat catchment, changes in the sink depth parameter had a large influence on the estimated fractions of direct and indirect connectivity.

So far, we only reported on the fraction of indirectly connected per total connected area. In Table 6, we additionally report the fractions of total agricultural area connected directly, indirectly, and not at all to surface waters. On average, we estimate between 5.5% and 38% (mean: 28%) of the agricultural area to be connected directly, 13% to 51% (mean: 35%) to be connected indirectly, and 12% to 77% (mean: 37%) not to be connected to surface waters. However, the variation between the catchments is much larger than the variation of the Monte Carlo analysis.

Table 6: Fractions of directly, indirectly, and not connected agricultural areas in our study catchments. The first row represent the mean fraction over all catchments and Monte Carlo simulations. The second row represents the median of the median over all catchments per MC simulation. The third row represents the median of the median over all MC analyses per catchment. In brackets, the minimum and the maximum median are given.

Statistic	Fraction of directly connected agricultural area f_{dir}	Fraction of indirectly connected agricultural area f_{indir}	Fraction of not connected agricultural area f_{nc}	Fraction of indirectly per total connected area $f_{fracindir}$
Mean	28%	35%	37%	57%
Median per MC simulation	25% (5.5%; 38%)	38% (13%; 51%)	32% (12%; 77%)	58% (43%; 74%)
Median per catchment	26% (1.8%; 70%)	37% (12%; 60%)	35% (3.9%; 53%)	57% (21%; 97%)

Sensitivity analysis

To analyse which model parameters have the largest influence on our model results, we tested the local model parameter sensitivity on our benchmark model. The fraction of indirectly to total connected area reacts most sensitive to changes in the road carving depth parameter. The difference between the minimal and maximal fraction reported was 17%. Results were also sensitive to the parameters shortcut definition (14%) and sink depth (13%). Infiltration width (4.3%) and hedge infiltration (2.5%) had only a minor influence on the fraction reported (see Figure S22 and Figure S23).

Hydrological activity

Systematic differences in hydrological activity between directly and indirectly connected areas would have a major influence on the interpretation of our connectivity analysis. We therefore tested for such differences by calculating the distributions of slope and topographic wetness index on these areas.

The distributions of both, slope and topographic wetness index were very similar for directly, indirectly, and not connected areas (see Figure S25 and Figure S26). Only the slope of not connected areas was found to be slightly smaller than the slope of connected areas. Hence, we could not identify any systematic differences in the factors affecting hydrological activity between directly and indirectly connected areas.

Consequently, given the current knowledge, the proportions of direct and indirect surface runoff entering surface waters are expected to be equal to the proportions of directly and indirectly connected agricultural areas. Analogously, if other boundary conditions of pesticide transport remain unchanged, directly and indirectly transported pesticide loads are expected to be proportional to directly and indirectly connected crop areas.

2.3.2.2 Extrapolation to the national level

We created a model for extrapolating the results of our study areas to the national level, using area fractions of the national erosion connectivity model (NECM) (Alder et al., 2015) aggregated to the catchment scale as explanatory variables. The area fractions of the NECM were transformed such that they fit the area fractions of the local surface runoff connectivity model (LSCM) resulting from the Monte Carlo analysis in our study areas. The resulting dataset is called the national surface runoff connectivity model (NSCM). The NSCM provides a separate model for each of the 100 Monte Carlo runs of the LSCM. It is aggregated to the catchment scale and covers all catchments of the valley zones, hill zones and lower elevation mountain zones. The differences between the fitted NSCM and the LSCM were strongly reduced compared to the original NECM (see Figure 9). The root-mean-square error (RSME) on average reduced from 17% to 9.5% for directly connected fractions, from 12% to 7.6% for indirectly connected fractions, and from 18% to 7.6% for not connected fractions.

By combining the NSCM with land use data, we came up with an estimate of connected crop areas on the national scale. Half of the Swiss agricultural areas in the model region are crop areas (i.e. arable land, vineyards, orchards, horticulture) and therefore potential pesticide source areas. On average, twenty six percent of crop areas (13% of total agricultural area) are connected directly, 34% (17% of total agricultural area) indirectly, and 40% (20% of total agricultural area) not at all (details: Figure S27; MC simulation quantiles: Table S9; spatial distribution: Figure S30 to Figure S36). From the total connected crop area, 54% (between 47 and 60%) are connected indirectly.

These results are similar to those obtained for the 20 study areas. Mean fractions of directly and indirectly connected agricultural areas are a bit smaller in the national scale estimation than for the 20 study areas (-2.0%, and -1.9%), while the fraction of not connected agricultural area is a bit larger (+3%). The fraction of indirectly connected crop area per total connected crop area is slightly smaller (-2.6%).

To assess if the national erosion connectivity model (NECM) is different from the national surface runoff connectivity model (NSCM), we determined the 5% and 95% quantiles of the NSCM predictions (see Table S9). If a fraction of the NECM is outside of this range, we considered this as a significantly different model prediction that is not expected, given our field data.

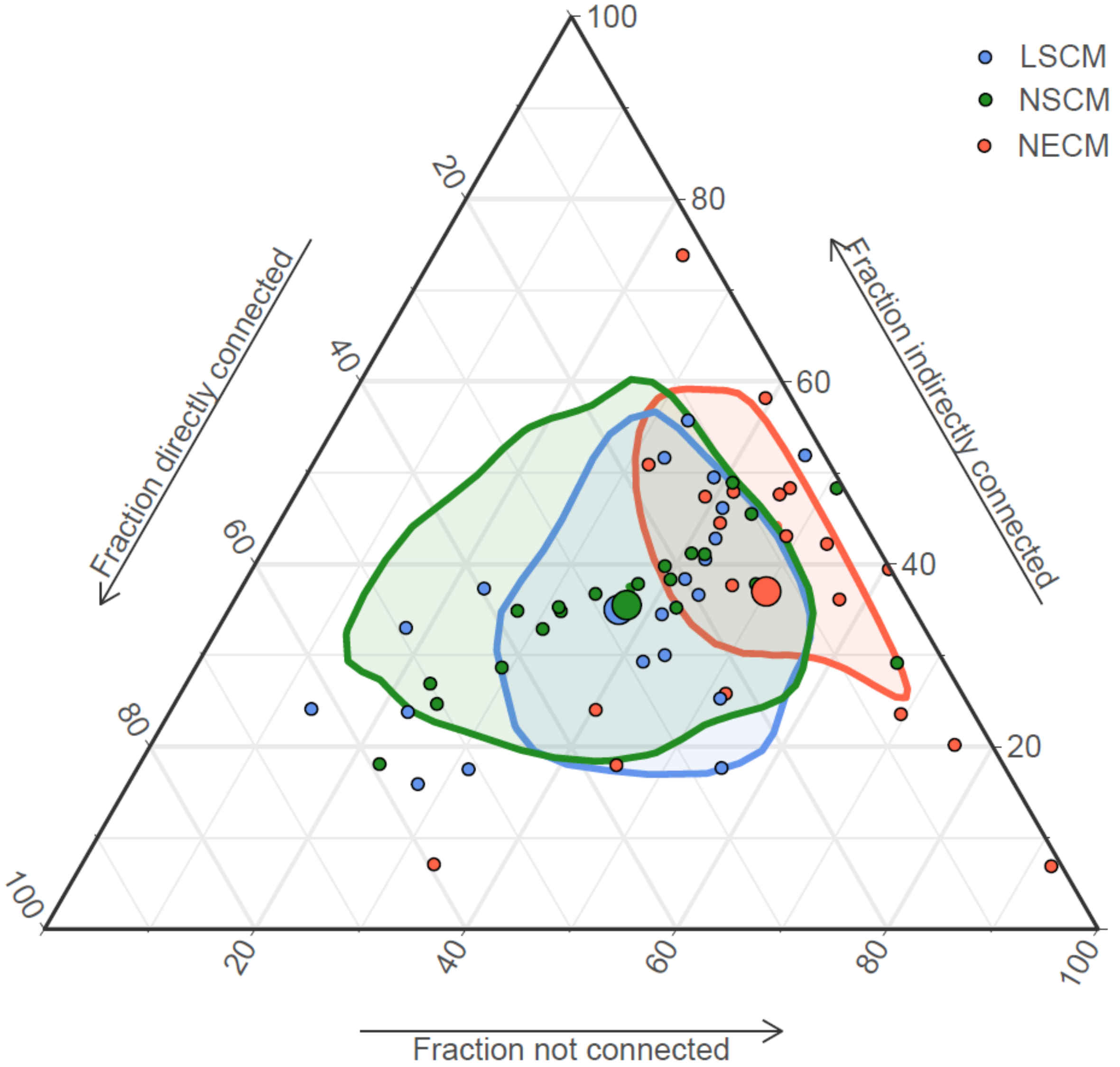


Figure 9: Fractions of directly connected (f_{dir}), indirectly connected (f_{indir}), and not connected areas (f_{nc}) per total agricultural area for the local surface runoff connectivity model (LSCM, blue), national erosion connectivity model (NECM, red), and national surface runoff connectivity model (NSCM, green) in the 20 study areas. Small blue circles represent the catchment medians of all Monte Carlo simulations of the LSCM, small red circles represent the data reported by the NECM, and small green circles represent the catchment medians of the NSCM. Large circles represent the means of the LSCM (blue), NECM (red), and NSCM data (green). Shaded areas represent normal Kernel density estimates of the LSCM, NECM, and NSCM data.

Compared to the NSCM, the NECM on average predicts lower fractions of directly connected crop areas $f_{crop,dir}$ (-6.4%), which is below the 5% quantile of the NSCM results. For indirectly connected areas $f_{crop,indir}$ (-0.9%), and not connected crop areas $f_{crop,nc}$ (+7.2%), the data reported by the NECM are within the 5% and 95% quantile of the NSCM results. However, the fraction of indirectly connected crop area per total connected crop area $f_{fracindir}$ reported by the NECM lies beyond the 95% quantile of the NSCM (+11%). In summary, $f_{crop,dir}$ and $f_{fracindir}$ reported by the NECM are significantly different from what would be expected from the NSCM. For $f_{crop,indir}$ and $f_{crop,nc}$, the reported fractions are in a similar range for both models. The results of the bootstrap (Figure S28) show that the differences between the two models are significantly larger than the uncertainty introduced by the selection of the study catchments.

The average difference in predicted connectivity fractions of *agricultural* areas between the two models ($\Delta f = ((f_{NSCM,dir} - f_{NECM,dir}) + (f_{NSCM,indir} - f_{NECM,indir}) + (f_{NSCM,nc} - f_{NECM,nc}))/3$) is strongly variable in space. Large differences are mainly found in large valleys (e.g. the Aare, Alpenrhein, and Rhone valleys, and the valleys of Ticino) and in the region of Lake Constance (see Figure S40). However, when looking at the difference in average predicted connectivity fractions of *crop* areas ($\Delta f_{crop} = ((f_{NSCM,crop,dir} - f_{NECM,crop,dir}) + (f_{NSCM,crop,indir} - f_{NECM,crop,indir}) + (f_{NSCM,crop,nc} - f_{NECM,crop,nc}))/3$), large differences almost exclusively are found in a band of catchments with high crop densities spreading through the Swiss midland (see Figure 10).

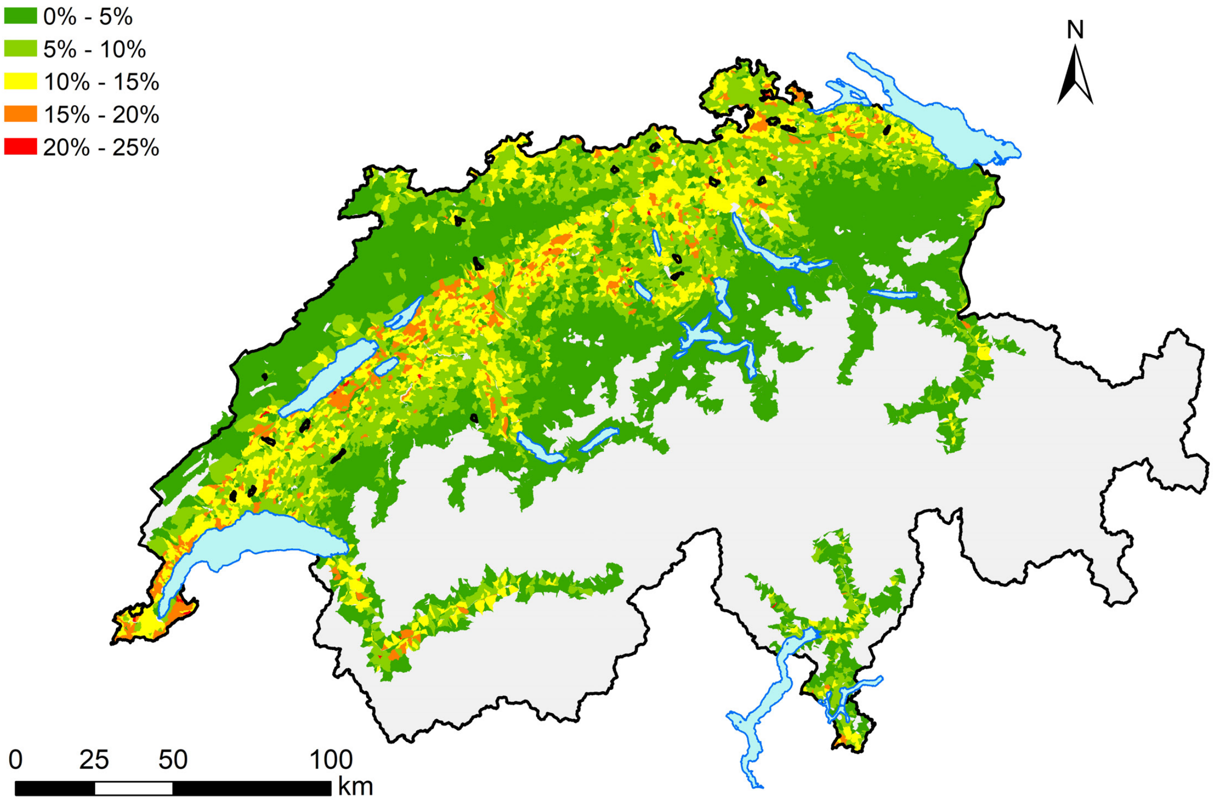


Figure 10: Average differences in connectivity fractions of crop areas between the NSCM and the NECM: $\Delta f_{crop} = ((f_{NSCM,crop,dir} - f_{NECM,crop,dir}) + (f_{NSCM,crop,indir} - f_{NECM,crop,indir}) + (f_{NSCM,crop,nc} - f_{NECM,crop,nc}))/3$. The map shows data for all Swiss catchments in the valley zones, hill zones and lower elevation mountain zones. Grey areas represent higher elevation mountain zones that were excluded from the analysis. Study areas are marked with black lines. Details on directly, indirectly, and not connected agricultural areas and crop areas are given in Figure S37 to Figure S43. For comparison, a map of crop densities is given in Figure S29. Source of background map: Swisstopo (2010)

2.4 Discussion

Occurrence of hydraulic shortcuts

Our study shows that storm drainage inlet shafts and maintenance shafts are common structures found in Swiss agricultural areas. While in neighbouring countries roads are often drained by ditches, Swiss roads are usually drained by storm drainage inlet shafts (Alder et al., 2015). It is therefore not surprising that most of the inlet shafts found in the study areas are located on roads. These findings are in accordance with the only other study in Switzerland reporting numbers on storm drainage inlet shafts (Prasuhn and Grünig, 2001).

The vast majority of mapped storm drainage inlet shafts were found to discharge to surface waters directly or via wastewater treatment plants (WWTPs). Thus, the occurrence of an inlet is in most cases directly related to a risk for pesticide transport to surface waters. The following three processes generate this risk: Firstly, pesticide loaded surface runoff produced on crop areas can enter the inlet. Secondly, spray drift deposited on roads can be washed off and enter the inlet. Thirdly, inlet shafts can be oversprayed during pesticide application, which is mainly considered probable for inlet shafts located in the fields.

Although maintenance shafts were also found to discharge to surface waters directly or via WWTPs, their occurrence does not directly translate into a risk for pesticide transport to surface waters. In contrast to storm drainage inlet shafts, maintenance shafts are not designed to collect surface runoff. Their lids are usually closed or only have a small opening, significantly decreasing the risk of surface runoff entering the shaft or of overspraying. In addition, lids of maintenance shafts in fields are often elevated compared to the soil surface. Maintenance shafts on roads are (in contrast to inlet shafts) usually positioned such that concentrated surface runoff is bypassing them. However, as also shown by Doppler et al. (2012), maintenance shafts can collect surface runoff from fields if they are located in a sink or a thalweg and water is ponding above them during rain events. During our field mapping campaign, we additionally found several damaged maintenance shafts that could easily act as a shortcut.

Channel drains and ditches discharging into surface waters were rare in most study areas with two exceptions. In Meyrin, the large length of these structures can be explained by the existence of a large vineyard. Additionally, the shaft density in this vineyard was higher than on the surrounding arable land. This indicates that vineyards could generally have higher shortcut densities than arable land. In Buchs, around 60% of the channel drain and ditch length consists of ditches that cannot be clearly distinguished from small streams. They are not appearing in the national topographic landscape model (Swisstopo, 2010) that was used for the definition of rivers and streams and did not appear to be streams during field mapping or when analysing aerial images.

The number of mapped shortcuts represents a lower boundary estimate of the shortcuts present (see results) and therefore leads to an underestimation of indirect connectivity. Probabilities for missing shortcuts during our mapping campaign depend on their location. While aerial images were at almost full coverage of the study areas, field mapping was performed mainly along roads. Drainage plans were available more often along roads than on fields. Therefore, we expect that detection probability of shortcuts is generally higher along roads than on fields. Besides coverage, various other factors influence the detection probabilities of the mapping methods. Field mapping and aerial image detection performance is reduced if shortcuts are covered. Along roads, this is mainly caused by leaves, soil, and for aerial images also by trees and vehicles. On the fields, this is mainly caused by soil or by crops. Detection performance of the aerial images method is additionally influenced by image quality and ground resolution. Image quality is mainly influenced by wind and light conditions during the UAV flights. In order to ensure high image quality, we planned UAV flights such that weather conditions were favourable (low wind, slightly overcast). However, differences in image quality between the study areas could not be completely avoided. Higher ground resolution could further improve the data produced. Although detection performance is not expected to be limited by the ground resolution used, higher resolution could improve the correct classification of shortcut types.

Surface runoff connectivity

Our study suggests that around half of the surface runoff connectivity in our study areas, but also on the national scale, is generated by hydraulic shortcuts. Surface runoff is considered one of the most important processes for pesticide transport to surface waters. Consequently, a large amount of the pesticide loads found in surface waters during rain events is expected to be transported by hydraulic shortcuts. These findings are in accordance to the results of other studies investigating the influence of hydraulic shortcuts on surface runoff connectivity (Alder et al., 2015; Bug and Mosimann, 2011; Prasuhn and Grünig, 2001) and on pesticide transport (Doppler et al., 2012).

The fraction of indirect connectivity was found to be very different between study areas. The variability introduced by the different properties of the study areas was larger than the variability introduced by the different model parameters of the Monte Carlo analysis, indicating that our results are robust against changes of our model parameters. Our model was most sensitive to changes of the parameters *road carving depth, shortcut definition*, and *sink depth*. These parameters are discussed in the following.

The parameter *road carving depth* accounts for the property of roads of collecting and concentrating surface runoff. This effect is strongly dependent on microtopography, extremely variable in space, and can therefore not be properly accounted for by a space-independent parameter. Usage of a higher resoluted digital elevation model could however reduce the uncertainty on the effect of roads on connectivity. Higher resolved digital elevation models could also help in capturing the influence of other microtopographical features better. For example, small ditches or small elevations on the ground can easily channel surface runoff. This can either direct surface runoff into a shortcut from areas not modelled to drain to a shortcut, or vice versa. In Switzerland, a new digital elevation model with a raster resolution of 0.5 m (Swisstopo, 2019a) recently became available and could be used for this purpose. This elevation model was not used within this study, since the study already had progressed further by the time the dataset was published.

The model parameters *shortcut definition* (i.e. are maintenance shafts in a sink considered as a shortcut) and *sink depth* are both related to the fate of surface runoff ponding in a sink. This indicates that maintenance shafts in sinks could have an important influence on surface runoff connectivity of agricultural areas. During our field mapping campaign, only few maintenance shafts in sinks were investigated. It is therefore unclear if most maintenance shafts in sinks are capturing ponding surface runoff, if surface runoff is usually infiltrating into the soil, or if it continues to flow on the surface. Sensitivity of our model to the parameter *sink depth* additionally indicates that sinks might play an important role for connectivity. Therefore, they should not be filled completely during GIS analyses, as this is done by default by some flow routing algorithms.

Surface runoff is usually assumed to drain to the receiving water of its topographical catchment. However, in various cases, the pipes draining hydraulic shortcuts were found to cross topographical catchment boundaries. Consequently, surface runoff and related pesticide loads are transported to a different receiving water than expected by the topographical catchment. This may be important to consider when interpreting pesticide monitoring data from small catchments. Similar effects were already reported for karstic aquifers or the storm drainage systems of urban areas (Jankowfsky et al., 2013; Luo et al., 2016).

Hydrological activity

We did not find any indication on systematic differences between the factors controlling hydrological activities of directly and indirectly connected agricultural areas by analysing slope and topographic wetness index. Those variables are a proxy for surface runoff formation, soil moisture, groundwater level, but also physical properties of the soil (Ayele et al., 2020; Sorensen et al., 2006). However, the hydrological activity of an agricultural area also depends on other factors that were not quantitatively analysed, such as *rainfall intensities, crop types, soil management practices*, or the presence of *tile drainage systems.*

Rainfall intensities: Because of the small size of the study areas and the close proximity between directly and indirectly connected areas, systematic differences in rainfall intensities within a catchment can be excluded.

Crop types and soil management can have a strong impact on runoff formation. These practices are chosen by the farmers and there could be systematic differences of these variables. For example, farmers aware of the effect of surface runoff and erosion on the pollution of surface waters might use different cultivation methods or crops (e.g. conservation tillage) on fields close to surface waters than on fields far away. This would lead to a higher probability of surface runoff formation on indirectly connected areas compared to directly connected areas. However, different cultivation methods require different farm machinery. Therefore, cultivation methods are often constrained by the machinery available and farmers use the same cultivation method per crop for all of their fields. Consequently, systematic differences in crop types or soil management between directly and indirectly connected areas of a catchment are unlikely.

Tile drainage systems: Shafts found in the field often belong to a tile drainage system. Therefore, fields on which shafts are located, have a higher probability to be drained by tile drainage systems than other fields. This could lead to higher infiltration capacities and consequently to reduced surface runoff on indirectly connected areas compared to directly connected areas. However, since most shafts are located along roads (see results) such differences would only have a minor effect on the overall surface runoff connectivity.

Although rainfall intensities, crop types, or soil management practices, are not expected to differ systematically within a catchment, they do differ across catchments. As mentioned in the results, we therefore expect the proportion of directly connected areas to indirectly connected areas in a catchment to be a good indicator for the proportion of surface runoff formed on directly and indirectly connected areas in this catchment. However, due to differences in hydrological activity, two catchments with similar total connected areas may differ strongly in the total amount of surface runoff formed.

Extrapolation to the national level

A major source of uncertainty in the national erosion connectivity model (NECM) is the usage of generalising assumptions due to lack of empirical data. Our results show that some of the estimated connectivity fractions of crop areas change significantly, when the NECM is transformed based on additional empirical data from our field study. However, the results of both models still are in the same order of magnitude and lead to the same general conclusion: At the national level, more than half of the connected crop area is connected to surface waters via hydraulic shortcuts, as we observed for the 20 study catchments. As shown in the results, large differences between the NECM and the NSCM in the predictions of crop area connectivity are almost exclusively found in one band of catchments with high cropping densities in the Swiss midland. Potential further empirical investigations or improvements of the NECM should therefore focus on a better representation of these catchments.

However, it is important to note, that within this study none of the models (NECM, LSCM, and NSCM) has been tested and validated empirically with independent data regarding their actual capacity to quantify the connectivity effects on surface runoff and related pesticide transport. These models provide predictions given the current availability of empirical observations. Suggestions for validating these models are given in the "further research" section.

From all tested variables, the NECM connectivity fractions showed the strongest correlations to the connectivity fractions reported by the local connectivity model (LSCM) in our study areas. This suggests that the NECM is a useful tool for assessing potential pesticide connectivity in relative terms (e.g. which catchments have high indirect connectivity compared to other catchments). Therefore, we recommend continuing to use the NECM in practice, e.g. as a starting point for identifying "hotspot" catchments of direct or indirect connectivity. Since the model results are not validated with independent data, they should always be combined with a verification in the field.

For creating the NSCM, all crop areas on which pesticides are commonly applied (arable land, vineyards, orchards, horticulture) were assumed to contribute by the same amount to the pesticide transport via surface runoff. However, these crop types are known to differ in the amounts of pesticide applied (De Baan et al., 2015), in the amounts of surface runoff produced, and also with respect to their connectivity to surface waters. This assumption could therefore be refined by considering pesticide application data and by investigating surface runoff connectivity in vineyards, orchards and horticulture in more detail.

Relevance in a broader geographical context

This study focussed on the relevance of hydraulic shortcuts in Switzerland. To our knowledge, no studies have systematically analysed the occurrence of hydraulic shortcuts in other countries. Nevertheless, the available literature suggests that in some regions such artificial structures like roads, pipes, or ditches are important for connecting fields with the stream network. For example, this was reported in the regions Alsace (FR) (Lefrancq et al., 2013), Lower Saxony (DE) (Bug and Mosimann, 2011), Baden-Wuerttemberg (DE) (Gassmann et al., 2012), or Rhineland-Palatinate (DE) (Rübel, 1999). Based on our findings, we hypothesise that shortcuts are mainly important in areas with small field sizes. This increases the density of linear structures such as roads for access.

Implications for practice

In Swiss plant protection[1] legislation and authorisation, the effect of hydraulic shortcuts on pesticide transport is currently not considered. Pesticide application is prohibited within a buffer of 3 m along open water bodies and according to the Swiss proof of ecological performance (PEP) vegetated buffer strip have to at least 6 m wide. In contrast, along roads, a buffer of only 0.5 m is required. Hence, the current Swiss legislation is protecting surface waters against direct, but not against indirect transport. This contrasts with the results of this study, suggesting that approximately half of the surface runoff related pesticide transport is occurring indirectly. This implies that there is evidence of a systematic gap in understanding and regulating pesticide risk at the national scale. The same gap was already pointed out by Alder et al. (2015) for soil erosion. However, beyond anecdotal evidence (e.g. Doppler et al. (2012)), this gap has not yet been validated with independent measurements of surface runoff and pesticide transport in the field.

While there remain important scientific questions about the validation of the suggested gap, authorities may wish to decide on mitigation measures despite such uncertainties. We therefore elaborate on potential mitigation measures in the following.

The most evident measure based on the current legislation are vegetated buffer strips along drained roads and around hydraulic shortcuts, infiltrating surface runoff before it reaches a shortcut. Generally, measures increasing infiltration capacity on the field would reduce pesticide transport. Other measures could aim on the shortcut structures themselves (e.g. construction of shortcuts as small infiltration

[1] In this study, we have been using the general term “pesticides” instead of “plant protection products” to make the text more readable. Since we only looked at substances used for plant protection in an agricultural context, the term “plant protection products” would have been more precise. The term “pesticides”, however, also includes “biocides” which are substances for control of plants or animals used in a non-agricultural context and were not subject of this study. The substances addressed in this study are regulated in the Swiss plant protection legislation and authorisation.

basins, removal of shortcuts, or treatment of water in shortcuts) or on the pipe outlets (e.g. drainage of shortcuts to infiltration basins, treatment of water at the pipe outlet).

Finally, pesticide transport via hydraulic shortcuts could be incorporated into the registration procedure and be considered for the mandatory mitigation measures that go with a registration. Models used in this context are currently only considering transport via direct surface runoff, erosion, tile drainages, and spray drift (De Baan, 2020).

Further research

Model validation. The model estimations presented here can give insight on pesticide transport via hydraulic shortcuts on the catchment and the national scale. However, as pointed out above, these models lack a field validation with independent measurements on flow and pesticide transport. In the following, we suggest validation approaches to overcome this limitation.

In our opinion, a validation of the local surface runoff connectivity model is ideally performed by measuring runoff and pesticide transport in a set of different small catchments. This should be done along a gradient of ratios between indirectly to directly connected areas (see Figure 8). Ideally, the catchments should be similar with respect to their structure (e.g. size, stream length, slope, land use, climate, or soil properties). Signals measured at the catchment outlet are always a superposition of different flow pathways. Therefore, runoff and pesticide transport through hydraulic shortcuts cannot be directly measured at the catchment outlet. To disentangle transport through hydraulic shortcuts from other pathways we foresee two different approaches.

The first approach aims on observing flow and transport within a catchment at locations where an unambiguous differentiation between the flow paths is possible. For example, hydraulic shortcuts in a catchment could be equipped with a discharge measurement and a water sampler. Such a setup would allow to determine the proportion of total catchment runoff and pesticide load that is transported via hydraulic shortcuts. In addition, isotopic tracers and runoff separation techniques could be used to determine the total amount of surface runoff contributing to catchment runoff. If the model is valid, the ratio of measured direct to measured indirect surface runoff should be proportional to the ratio of directly to indirectly connected areas. Additionally, these measurements could be used to improve the parametrisation of the local connectivity model.

However, due to the large numbers of measurement locations needed, the above-mentioned validation approach would be very laborious. The second validation approach therefore aims on disentangling transport through hydraulic shortcuts while only measuring at the catchment outlet of a set of catchments. For the interpretation of the local connectivity model, we assumed that direct and indirect surface runoff are proportional to the directly and indirectly connected area. If this assumption is valid, more surface runoff should reach the stream in catchments with larger fractions of connected areas. Consequently, in such catchments, runoff coefficients should be higher during discharge events that are predominenantly triggered by Hortonian overland flow such as intensive thunderstorms. For these events, uncertainties introduced by different subsurface properties of the catchments play a minor role compared to other events. Furthermore, if a set of catchments has similar fractions of directly connected area, but different fractions of indirectly connected area, larger runoff coefficients should be measured in catchments with larger fractions of indirectly connected area.

If the local connectivity model proves valid on the catchment scale, the question would be how to improve on the spatial extrapolation to the national scale. Except for the occurrence of hydraulic shortcuts, all input data for the local connectivity model are available on this larger scale as well. Therefore, the local connectivity model can easily be extended to much larger scales if the occurrence of hydraulic shortcuts is known. However, the shortcut mapping procedure used in this study is time-consuming. Thus, to efficiently map shortcuts on larger scales, automated algorithms for inlet localization using remote sensing data could be used (e.g. Mattheuwsen and Vergauwen (2020), Moy de Vitry et al. (2018)). An application of the local connectivity model to larger scales could then replace the extrapolation approach used in this study, eliminating the associated uncertainty.

Shortcuts in vineyards. Our results (i.e. Meyrin and additional field observations) suggest that the presence of hydraulic shortcuts as well as the fraction of indirectly connected areas are higher in vineyards than on arable land. Since this study focused mainly on the latter, the sample size was too small for a quantitative analysis of vineyards. The fact that Swiss vineyards usually have high road densities points into the same direction. In Swiss vineyards, pesticides are applied more often and in larger amounts than on arable land (De Baan et al., 2015). Therefore, an assessment of hydraulic shortcut relevance in vineyards is needed.

Spray drift on roads. Hydraulic shortcuts are not only collecting surface runoff from target areas, but also from non-target areas such as roads. As shown by Lefrancq et al. (2013), large amounts of spray drift can be deposited on roads. These deposits are expected to be washed off during rain events and to be transported to surface waters via hydraulic shortcuts. Further research is needed to quantify the relevance of this process for pesticide pollution in streams.

Hydrological activity. In our discussion on the hydrological activity (see above), we explained that systematic differences in hydrological activity are unlikely within a catchment, but are expected across catchments. Further research should aim on quantifying the differences in hydrological activity across catchments and their influence on runoff formation. Some of the datasets that could serve such a comparison are available on the national scale (e.g. map of tile drainage potential (Koch and Prasuhn, 2020), or rainfall statistics (e.g. Frei et al. (2018)). Other datasets are currently being developed (e.g. a national plot-specific crop type dataset) or have to be developed (e.g. national soil maps).

2.5 Conclusions

Our study shows that hydraulic shortcuts are common structures found in Swiss arable land areas of the Swiss plateau. Shortcuts are found mainly along roads, but also directly in the field. The analyses suggests that on average, around half of the surface runoff connectivity on Swiss arable land is caused by hydraulic shortcuts. Further analyses on hydrological activity and crop density suggest that the same proportion of surface runoff and related pesticide load is transported to surface waters through hydraulic shortcuts. This statement holds for both, the selected study catchments, and the whole country. However, in Swiss pesticide legislation and pesticide authorisation, hydraulic shortcuts are currently not considered. Therefore, current regulations may fall short to address the full extent of the problem.

The field data acquired in this study suggest that the national erosion connectivity model (NECM) is a useful tool for relatively comparing potential pesticide connectivity between catchments. However, the results also show that additional field data significantly changed the reported connectivity fractions and improved the model reliability.

Overall, the findings highlight the relevance of better understanding the connectivity between fields and the receiving water, as well as the underlying factors and physical structures in the landscape. The model results of this study lack a validation with field measurements on actual water flow and pesticide transport in hydraulic shortcuts. This should be addressed in further research. Propositions for such validations are presented in the discussion section.

This study focused on the contribution of hydraulic shortcuts to surface runoff connectivity and related pesticide transport on arable land. However, for other crop types, the contribution of shortcuts is expected to be different. Especially in vineyards, we expect a higher contribution due to their spatial structure (e.g. high road densities, or steep slopes) and due to higher pesticide use.

Chapter 3. Are spray drift losses to agricultural roads more important for surface water contamination than direct drift to surface waters?

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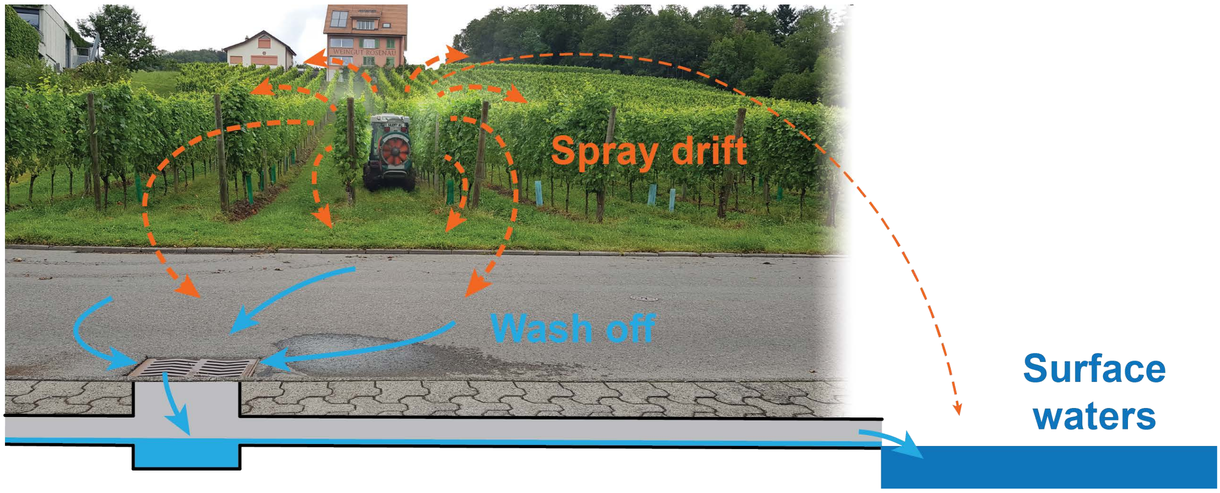
Graphical abstract



Abstract

Spray drift is considered a major pesticide transport pathway to surface waters. Current research and legislation usually only considers direct spray drift. However, also spray drift on roads and subsequent wash-off to surface waters was identified as a possible transport pathway. Hydraulic shortcuts (storm drainage inlets, channel drains, ditches) have been shown to connect roads to surface waters, thus increasing the risk of drift wash-off to surface waters. However, the importance of this pathway has never been assessed on larger scales. To address this knowledge gap, we studied 26 agricultural catchments with a predominance of arable cropping (n=17) and vineyards (n=9). In these study sites, we assessed the occurrence of shortcuts by field mapping. Afterwards, we modelled the areas of roads drained to surface waters using a high-resolution digital elevation model (0.5 m resolution) and a multiple flow algorithm. Finally, we modelled drift deposition to drained roads and surface waters using a spatially explicit, georeferenced spray drift model. Our results show that for most sites, the drift to drained roads is much larger than the direct drift to surface waters. Assuming that farmers comply with the legally required buffer distances, drift to roads exceeds the direct drift by a factor of 4.5 to 18 in arable land sites, and by 35 to 140 in vineyard sites. In arable land sites, drift to drained roads is rather small (0.0015% to 0.0049% of applied amount) compared to typical total pesticide losses to surface waters. However, substantial drift to drained roads in vineyard sites was found (0.063% to 0.20% of applied amount). Current literature suggests that major fractions of the drift deposited on roads can be washed off during rain events, especially for pesticides with low soil adsorption coefficients. For such pesticides and particularly in vineyards, spray drift wash-off from drained roads is therefore expected to be a major transport pathway to surface waters.

3.1 Introduction

After pesticide application on agricultural crops, a certain fraction of the applied amount is not attained to the target crop, but is lost to non-target ecosystems such as surface waters. These pesticide losses pose a major threat for aquatic ecosystems (Schwarzenbach et al., 2006; Stehle and Schulz, 2015). Besides point sources (e.g. farmyard runoff, accidental spills, combined sewer overflows, or and wastewater treatment plants), surface runoff (Larsbo et al., 2016; Reichenberger et al., 2007), macropore flow to tile drainages (Kladivko et al., 2001; Sandin et al., 2018), and spray drift (Carlsen et al., 2006; Ganzelmeier, 1995) are considered the most important pesticide transport pathways to surface waters. Spray drift is defined as the downwind movement of spray droplets beyond the target area of application originating from the spraying process (Stephenson et al., 2006). Studies quantifying surface water pollution by spray drift are typically only considering drift directly deposited on surface waters. This holds for modelling studies (Huber et al., 2000; Padovani et al., 2004; Röpke et al., 2004; Travis and Hendley, 2001), field studies (Bonzini et al., 2006; Schulz, 2001), and is also the case for the models used in the European pesticide authorisation (Linders et al., 2003). However, spray drift is also deposited on various other non-target areas (e.g. soils, non-target crops, forests, settlements, roads, farm tracks). Depending on the spraying device, the non-target deposition is estimated to 0.8 – 4% of the applied amount for ground applications (Jensen and Olesen, 2014; Viret et al., 2003). Depending on the properties of the non-target area, some of this spray drift may be washed off to surface waters during subsequent rainfall events (Gassmann et al., 2013; Schönenberger et al., 2022a).

Roads and farm tracks have a very low infiltration capacity and limited sorption potential (Ramwell, 2005). Therefore, on these areas, surface runoff is formed with higher frequency and pesticides are washed off much easier than from target areas or from other non-target area types. Especially substances with low soil adsorption coefficients (K_{oc} < 250 mL g^{-1}) have been shown to be washed off in large amounts (57% or more of the applied amount) during simulated and natural rainfall (Ramwell et al., 2002; Thuyet et al., 2012). However, also for substances with higher K_{oc}, relevant wash-off fractions have been reported during the first rainfall after application, e.g. up to 5.8% (Thuyet et al., 2012) and up to 2.7% (Jiang et al., 2012).

Roads and farm tracks in agricultural areas are often drained by storm water drainage inlets or by other artificial structures (e.g. channel drains or ditches) (Alder et al., 2015; Payraudeau et al., 2009; Rübel, 1999; Schönenberger and Stamm, 2021). Especially in Switzerland, these structures are often connected to surface waters via subsurface pipe systems. This enables spray drift wash-off from remote roads to reach surface waters and therefore creates a so-called shortcut (Doppler et al., 2012). These shortcuts therefore strongly increase the potential of spray drift wash-off from roads for surface water pollution.

Despite its large potential for pesticide transport to surface waters, only in four catchments measurements providing insights on this transport process were performed to the best of our knowledge: In a German vineyard catchment, Rübel (1999) found that drift on vineyard roads during helicopter applications was leading to high pesticide concentrations in the receiving stream in the following rain event. Ground applications were found to have a similar effect, but were leading to lower maximal concentrations compared to helicopter spraying. In a French vineyard catchment, Lefrancq et al. (2014) reported spray drift on roads and subsequent wash-off to be responsible for a large fraction of the runoff-related fungicide load at the catchment outlet. In a Swiss arable land catchment, Schönenberger et al. (2022a) found that either spray drift on roads or droplet losses from the spraying equipment led to increased pesticide concentrations in inlets of the road storm water drainage system. Finally, in another Swiss arable land catchment, Ammann et al. (2020) found – based on the field study described in Doppler et al. (2012) – that the consideration of spray drift wash-off from roads could strongly reduce the uncertainty of exposure models.

These studies show that spray drift wash-off from roads is a relevant transport pathway to surface waters in certain catchments. However, it remains unclear how much spray drift is deposited on roads draining to surface waters for larger spatial scales, and how the amount deposited differs between catchments and crop types. In addition, it is unknown to which degree drift reduction measures could reduce pesticide losses caused by this pathway.

For assessing spray drift to surface waters (usually streams, but also ditches or ponds) on larger spatial scales, various studies have applied spatially explicit georeferenced drift models (Holterman and Van de Zande, 2008; Kubiak et al., 2014; Schad and Schulz, 2011; Wang and Rautmann, 2008). These models combine spatial data on surface waters and sprayed crops with spray drift deposition functions obtained from experimental trials (Ganzelmeier, 1995; Rautmann et al., 1999). To our knowledge, however, they have never been applied to determine drift deposition on non-target areas other than surface waters, particularly not for roads or farm tracks.

In this study, we therefore aimed at comparing spray drift deposition on surface waters to the deposition on roads and farm tracks draining to surface waters. For this, we combined a field mapping approach with a spatially explicit, georeferenced spray drift model for a large set of agricultural catchments representing arable land and vineyards in Switzerland. We focused on these two crop types since they are two of the most important crop types in Switzerland with respect to coverage (arable land) and average pesticide use (vineyards). Additionally, spray drift deposition differs strongly between those two crop types due to different spraying methods (boom sprayers on arable land, and air blast sprayers on vineyards), and different spatial structures (e.g. density and size of roads around crop areas) (Schönenberger and Stamm, 2021). In some Swiss vineyard regions, also helicopters are still used for spraying. This method is however not addressed in this study.

Our research questions are:

- How much spray drift is deposited on roads and farm tracks draining to surface waters? In comparison, how much spray drift is deposited in surface waters directly?
- How do the deposited amounts differ between arable land and vineyards?

- How much would the drift on drained roads and farm tracks be reduced by spray drift buffers?

Based on the respective results, we also aim at answering the question how important spray drift wash-off from roads may be for the pesticide pollution of surface waters compared to direct spray drift, and compared to total pesticide losses. However, given the paucity of empirical data on wash-off from these surfaces the results will be only tentative at this stage.

3.2 Material and Methods

3.2.1 Selection of study sites

We selected two sets of agricultural catchments as study sites for our analysis. One set represents Swiss arable land areas, the other one Swiss vineyards. The arable land and vineyard sites were selected randomly from a nationwide, small-scale topographic catchment dataset (BAFU, 2012). The selection probability of each catchment equalled the arable land and vineyard area in the catchment, respectively, as reported by the Swiss land use statistics (BFS, 2014) (details – Schönenberger and Stamm (2021) for arable land, Simon (2019) for vineyards). From the resulting sites (20 arable land, 8 vineyards), we removed four sites for which no high-resolution crop data were available. Additionally, two vineyard sites only consisted of small-scale plots in settlements. Since this type of small-scale viticulture is a special case present only in few areas, these two sites were also removed. In contrast to the other sites, the site Meyrin contains both, large arable land areas and large vineyards. This site was therefore splitted into an arable land part and a vineyard part (see Table 7). Finally, the selected sites were complemented by three catchments used in previous studies assessing pesticide concentrations in surface waters (Schönenberger et al., 2022a; Spycher et al., 2018; Spycher et al., 2019). The resulting 26 sites (17 arable land, 9 vineyards) are shown in Figure 11.

3.2.2 Modelling procedure

For this study, we considered two types of non-target areas: Surface waters and drained roads. Other non-target areas (e.g. hedges, plot margins) were considered irrelevant for subsequent transfer to surface waters since surface runoff formation on these areas is rare compared to roads (see Sect. 3.1). Drained roads were defined as roads from which water drains to surface waters while only flowing along roads or through shortcuts. They were categorized into roads drained to surface waters via shortcuts and into roads directly draining to surface waters. For determining drained roads, we first mapped shortcuts in the study catchments and then combined these maps with a surface runoff connectivity model (see Figure 12). Afterwards, we determined the amount of spray drift deposited on drained roads and on surface waters using a spray drift model. In the following, these steps are described in detail.

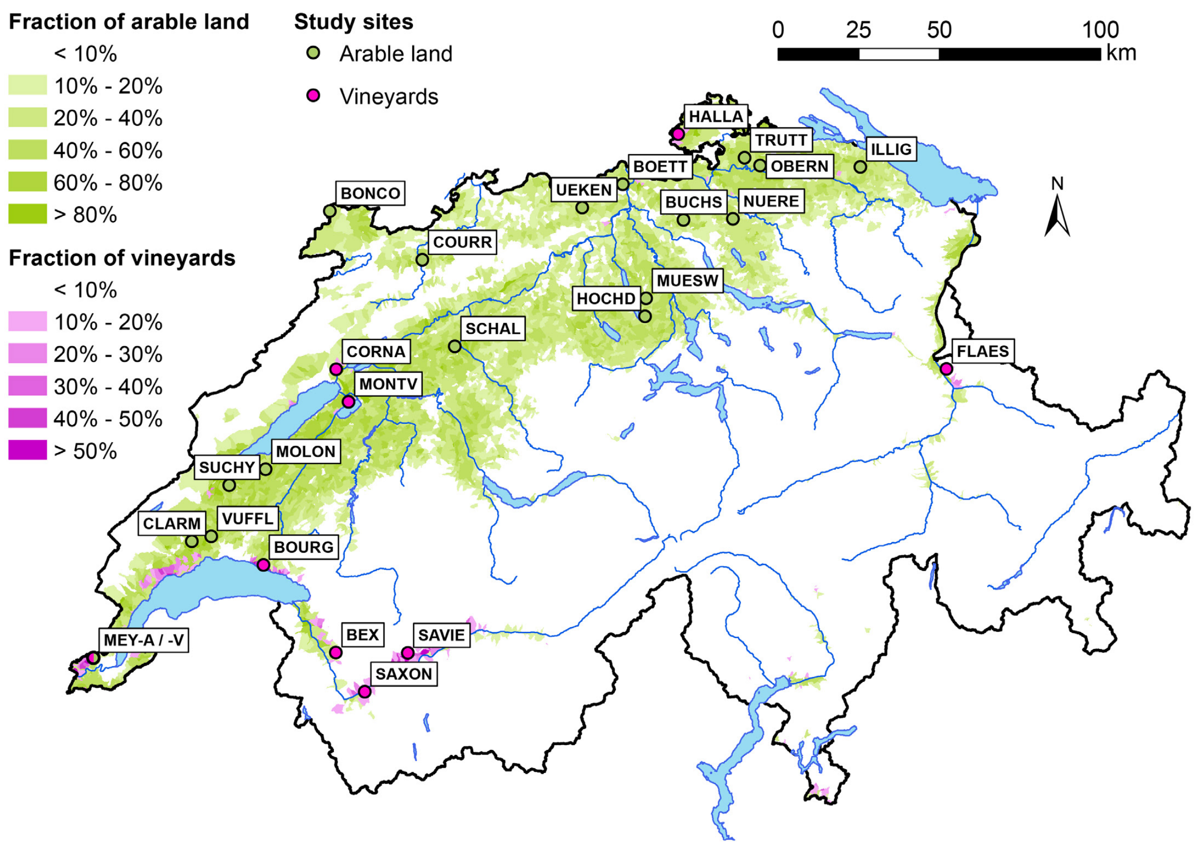


Figure 11: Locations of the study sites and fractions of arable land and vineyards in Swiss hydrological catchments. Sources: BAFU (2012), BFS (2014), Swisstopo (2010).

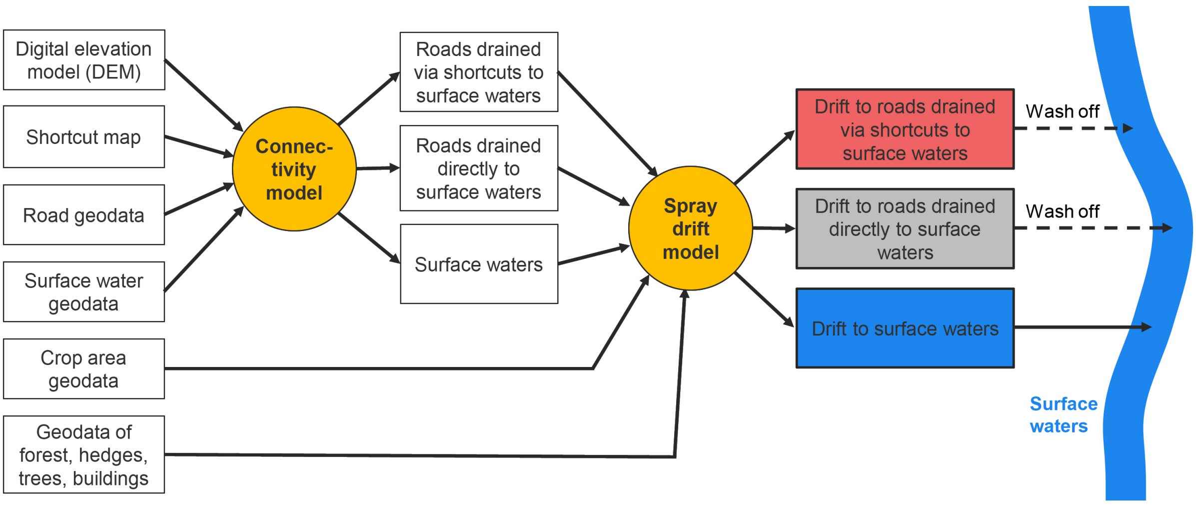


Figure 12: Schematic representation of the procedure for modelling spray drift to roads drained via inlets, to roads drained directly to surface waters, and to surface waters.

3.2.2.1 Mapping shortcuts

Shortcuts were defined as artificial structures increasing and/or accelerating the process of surface runoff reaching surface waters (Schönenberger and Stamm, 2021). Within this study, we considered storm drainage inlet shafts, channel drains, and ditches along roads and farm tracks as potential shortcuts. These potential shortcuts were defined as real shortcuts, if they are drained to surface waters, to wastewater treatment plants or to combined sewer overflows.

In all 26 study sites, we mapped potential shortcuts along roads and farm tracks. For the arable land sites, mapping was performed in 2017 and 2018 for the whole catchments, combining three different methods: Field surveys, storm drainage system plans, and high resolution aerial images (resolution: 2.5 to 5 cm) from an unoccupied aerial vehicle (details – Schönenberger and Stamm (2021)). For vineyard sites, mapping was performed in 2019. In contrast to arable land, which is often distributed throughout the whole catchment, vineyards usually only cover a certain part of the catchment. We therefore did not map potential shortcuts in the whole catchment, but only along roads and farm tracks adjacent to vineyards. The mapping in vineyards was based on field surveys (details – Simon (2019)), complemented with data from storm drainage system plans, Google Street View, and aerial images with intermediate resolution (10 cm) (Swisstopo, 2019b).

Table 7: Overview over study sites. Selection: R – random selection, M – site used in previous studies

ID	Study site	Canton	Abbreviation	Crop type	Selection	Area (km^2)
1	Böttstein	AG	BOETT	Arable	R	3.34
2	Boncourt	JU	BONCO	Arable	R	5.90
3	Buchs	ZH	BUCHS	Arable	R	3.86
4	Clarmont	VD	CLARM	Arable	R	2.47
5	Courroux	JU	COURR	Arable	R	2.80
6	Hochdorf	LU	HOCHD	Arable	R	2.37
7	Illighausen	TG	ILLIG	Arable	R	1.90
8	Molondin	VD	MOLON	Arable	R	4.15
9	Müswangen	LU	MUESW	Arable	R	3.00
10	Nürensdorf	ZH	NUERE	Arable	R	2.34
11	Oberneunforn	TG	OBERN	Arable	R	3.30
12	Schalunen	BE	SCHAL	Arable	M	2.78
13	Suchy	VD	SUCHY	Arable	R	3.28
14	Truttikon	ZH	TRUTT	Arable	R	5.06
15	Ueken	AG	UEKEN	Arable	R	1.99
16	Vufflens-la-Ville	VD	VUFFL	Arable	R	2.79
17	Meyrin (arable)	GE	MEY-A	Arable	R	8.50
18	Bex	VD	BEX	Vineyard	R	4.27
19	Bourg-en-Lavaux	VD	BOURG	Vineyard	R	0.67
20	Cornaux	NE	CORNA	Vineyard	R	2.76
21	Fläsch	GR	FLAES	Vineyard	R	2.29
22	Hallau	SH	HALLA	Vineyard	M	0.98
23	Meyrin (vineyard)	GE	MEY-V	Vineyard	R	1.50
24	Mont-Vully	FR	MONTV	Vineyard	R	1.63
25	Savièse	VS	SAVIE	Vineyard	M	2.41
26	Saxon	VS	SAXON	Vineyard	R	4.25
				Average		**3.22**

If storm drainage plans were available for the respective study site, we additionally determined where potential shortcut structures drain to. Structures draining to surface waters, wastewater treatment plants and/or combined sewer overflows were considered as real shortcuts. Structures draining to infiltration areas (e.g. infiltration ponds, forests, or grassland) were not considered as real shortcuts and were neglected in the further steps. Ninety-nine percent of the storm drainage inlets, and 98% of the channel drains and ditches were found to be real shortcuts in a previous study (Schönenberger and Stamm, 2021). Therefore, potential shortcuts for which no drainage plans were available were assumed to act as real shortcuts.

3.2.2.2 Surface runoff connectivity model

To determine drained roads in the study sites, we used a modified version of the surface runoff connectivity model described in Schönenberger and Stamm (2021). This was done in four steps as described below. How the required model parameters were chosen and how their influence on the model results was assessed, is described in Sect. 3.2.2.4.

1) *Determination of road areas.* Road and farm track areas (called road areas in the following) were derived from the topographical landscape model swissTLM3D (Swisstopo, 2020b), and were complemented with other sealed areas from the same dataset (parking lots, motorway stations). Details are given in S3.1.1.
2) *Determination of surface water areas.* Surface water areas (streams and stagnant waters) were also derived from the topographical landscape model swissTLM3D (Swisstopo, 2020b). Details are given in S3.1.2.
3) *Determination shortcut areas.* Shortcut areas were defined as the area 1 m around the mapped shortcut structures.
4) *Determination of connectivity.* For determining the connectivity of road areas to surface water or shortcut areas, we used the TauDEM model (Tarboton, 1997), which is based on a D-infinity flow direction algorithm. As an input, we used a digital elevation model (DEM) with a resolution of 0.5 m (Swisstopo, 2020a) that was modified as follows. Firstly, to account for the surface runoff accumulation effect of roads (Fiener et al., 2011; Heathwaite et al., 2005), road areas were carved into the DEM by a certain depth (parameter *road carving depth* d_{road}). Secondly, all topographic sinks smaller than a certain depth (parameter *sink fill depth* d_{sink}) were filled. Finally, surface water areas and shortcut areas were carved 50 m and 20 m into the DEM. These large carving depths ensured that raster cells representing surface water and shortcut areas were much lower than the surrounding terrain. This guaranteed that the flow direction of the raster cells adjacent to surface water and shortcut areas pointed towards these areas. The modified DEM, shortcut areas, and surface water areas were then used as an input for the D-infinity upslope dependence tool of the TauDEM model. As a result, we obtained a raster containing all roads drained to surface waters or shortcuts. Some of the raster cells classified as

drained roads had a flow path running for longer distances over fields or through forests. However, we expect runoff formed on roads to infiltrate when flowing for longer distances on these areas. Therefore, we removed drained road cells from the raster dataset if their flow path outside roads exceeded a maximal distance (parameter *infiltration distance* d_{inf}).

To assess which area of drained roads per crop area is found per study site s and how this compares to the area of surface waters, we calculated drainage densities d_s (drained area per crop area) as follows:

$$d_s = \begin{pmatrix} d_{RSC,s} \\ d_{RSW,s} \\ d_{SW,s} \end{pmatrix} = \frac{\begin{pmatrix} A_{RSC,s} \\ A_{RSW,s} \\ A_{SW,s} \end{pmatrix}}{A_{crop,s}} \tag{3.1}$$

$d_{RSC,s}$, $d_{RSW,s}$, and $d_{SW,s}$ are the drainage density of roads drained to shortcuts, the drainage density of roads drained to surface waters, and the drainage density of surface waters in study site s. $A_{RSC,s}$, $A_{RSW,s}$, and $A_{SW,s}$ are the area of roads drained to shortcuts, of roads drained to surface waters, and of surface waters in study site s. $A_{crop,s}$ is the crop area in study site s.

3.2.2.3 Spray drift model

The spray drift model developed in this study determines drift from crop areas to the relevant non-target areas (i.e. drained roads and surface waters) based on their spatial arrangement in the study sites. Additionally, the model considers drift reduction by barriers, such as forest, hedges, trees, or buildings. In this section, we first describe how the input data were prepared, and afterwards how spray drift was modelled. In Sect. 3.2.2.4, we describe how model parameters were chosen and how the model uncertainty was assessed.

Input data. As input data for the spray drift model, we used the areas of drained roads and of surface waters, determined by the connectivity model (Sect. 3.2.2.2). Drained roads and streams were rasterized with a resolution of 2 x 2 m. Larger surface waters (e.g. ponds, lakes, large rivers) were rasterized with a resolution of 10 x 10 m. The areas of forest, hedges, trees, buildings, and vineyards were obtained from the topographical landscape model swissTLM3D (Swisstopo, 2020b). This dataset does however not specify the extent of arable land. Arable land areas were therefore extracted from a collection of standardized cantonal datasets on agricultural areas in parcel resolution (Kanton Aargau et al., 2020).

We assumed that pesticides are applied according to Swiss regulations and to Swiss proof of ecological performance (ChemRRV, 2005; DZV, 2013). These regulations prohibit pesticide applications within a buffer of 6 m around surface waters, 3 m around hedges, forests, and riparian woods, and 0.5 m around roads and farm tracks. For our analysis, we therefore removed all crop areas (vineyards, arable land) lying inside these buffers.

Spray drift model. The spray drift model developed in this study is based on spray drift curves according to Rautmann et al. (1999). They describe the spray drift deposition $\rho_{drift,i,p}$ (kg m^{-2}) on a non-target area i depending on its upwind distance $d_{i,p}$ (m) to a sprayed plot p (eq. 3.2). $\rho_{appl,p}$ is the application rate (kg m^{-2}) on the sprayed plot.

$$\rho_{drift,i,p} = a \cdot {d_{i,p}}^{b} \cdot \rho_{appl,p} \quad (3.2)$$

The curves were derived in field trials for wind speeds between 1 and 5 m/s. The minimal drift distances measured in the field trials were 1 m for arable land and 3 m for vineyards. The maximal distances were 100 m for both trial types. For this study, we extrapolated the drift curve to a minimal drift distance of 0.5 m, and to a maximal distance as defined by the parameter *maximal drift distance* $d_{drift,max}$.

The upwind distance from a non-target area cell to the next sprayed plot depends strongly on the wind direction. Similar to other studies (e.g. Wang and Rautmann (2008), Golla et al. (2011)), we therefore calculated the upwind distances for eight different wind directions (N, NE, E, SE, S, SW, W, NW). In the field trials used for the determination of spray drift curves, sprayed plots had a standardized width of around 20 m parallel to the wind direction (Julius Kühn-Institut, 2013). However, in the study sites analysed here, the extent of crop areas along the wind line was often larger than these 20 m. In these cases, we assumed the drift of these crop areas to equal the drift produced by a sequence of standard plots located in intervals of 20 m along the wind line (example – Figure 13). For each of these standard plots, drift was calculated separately, summed up, and multiplied with the area A_i (m^2) of the non-target area cell to determine the spray drift $m_{drift,i,w}$ (kg) for wind direction w to the non-target area cell i (eq. 3.3).

$$m_{drift,i,w} = \sum_{p=1}^{n_p} \left(a \cdot {d_{i,p.w}}^{b} \cdot \rho_{appl,p} \right) \cdot A_i \quad (3.3)$$

In the model, we additionally considered the interception of spray drift by barriers, such as forests, hedges, trees, or buildings. For forest, hedges, and trees (FHT), we assumed that drift is linearly reduced proportional to their distance $d_{FHT,i,p,w}$ (m) along the wind line (see Figure 13) between the sprayed plot and the non-target area. The amount of drift reduction is described by the drift reduction factor $f_{FHT,i,p,w}$ (eq. 3.4). The distance needed for intercepting all spray drift is described by the model parameter *distance of forest, hedges, or trees causing full drift interception* $d_{FHT,int}$ (m). An example of how $d_{FHT,i,p,w}$ is calculated if a FHT polygon is located further away of the non-target area is provided in Sect. S3.1.3.

$$f_{FHT,i,p,w} = \max\left(1 - \frac{d_{FHT,i,p,w}}{d_{FHT,int}}, 0\right) \quad (3.4)$$

Similarly, a drift reduction factor for buildings $f_{B,i,p,w}$ was added to the model (eq. 3.5). If a building is located between the sprayed plot and the non-target area, the spray drift is reduced as specified by the model parameter *spray drift interception by buildings* $f_{B,int}$. $d_{B,i,p,w}$ is the distance of buildings between the sprayed plot and the non-target area along the wind line.

$$f_{B,i,p,w} = \begin{cases} 1 \mid d_{B,i,p,w} = 0 \\ 1 - f_{B,int} \mid d_{B,i,p,w} > 0 \end{cases} \quad (3.5)$$

In a last step, we combined eq. 3.3 to 3.5. Assuming that the application rate ρ_{appl} is the same for all crop areas per study site, the amount of spray drift lost per total amount applied per study site $f_{lost,w}$ was calculated as shown in eq. 3.6. For each study site, the amount of spray drift lost per applied amount was calculated for all three non-target area types, and all eight wind directions. Additionally, we calculated the relative losses $f_{lost,rel}$ to each non-target area type (eq. 3.7; RSC – roads drained to shortcuts, RSW – roads drained to surface waters, SW – surface waters).

$$f_{lost,w} = \frac{\sum_{i=1}^{n_i} m_{drift,i,w}}{m_{appl}} = \frac{\sum_{i=1}^{n_i}\left(\sum_{p=1}^{n_p}\left(a \cdot d_{i,p,w}{}^{b} \cdot f_{FHT,i,p,w} \cdot f_{B,i,p,w}\right) \cdot A_i\right) \cdot \rho_{appl}}{A_{crop} \cdot \rho_{appl}} \qquad (3.6)$$

$$\begin{pmatrix} f_{lost,rel,RSC} \\ f_{lost,rel,RSW} \\ f_{lost,rel,SW} \end{pmatrix} = \frac{\begin{pmatrix} f_{lost,RSC} \\ f_{lost,RSW} \\ f_{lost,SW} \end{pmatrix}}{f_{lost,RSC} + f_{lost,RSW} + f_{lost,SW}} \qquad (3.7)$$

For the crop-specific drift parameters a and b, we used an updated version of the median spray drift parameters of Rautmann et al. (1999) provided by the authors of the publication. For arable land, they equalled 0.9658 (a) and -0.9507 (b), for vineyards 30.408 (a) and -1.5987 (b).

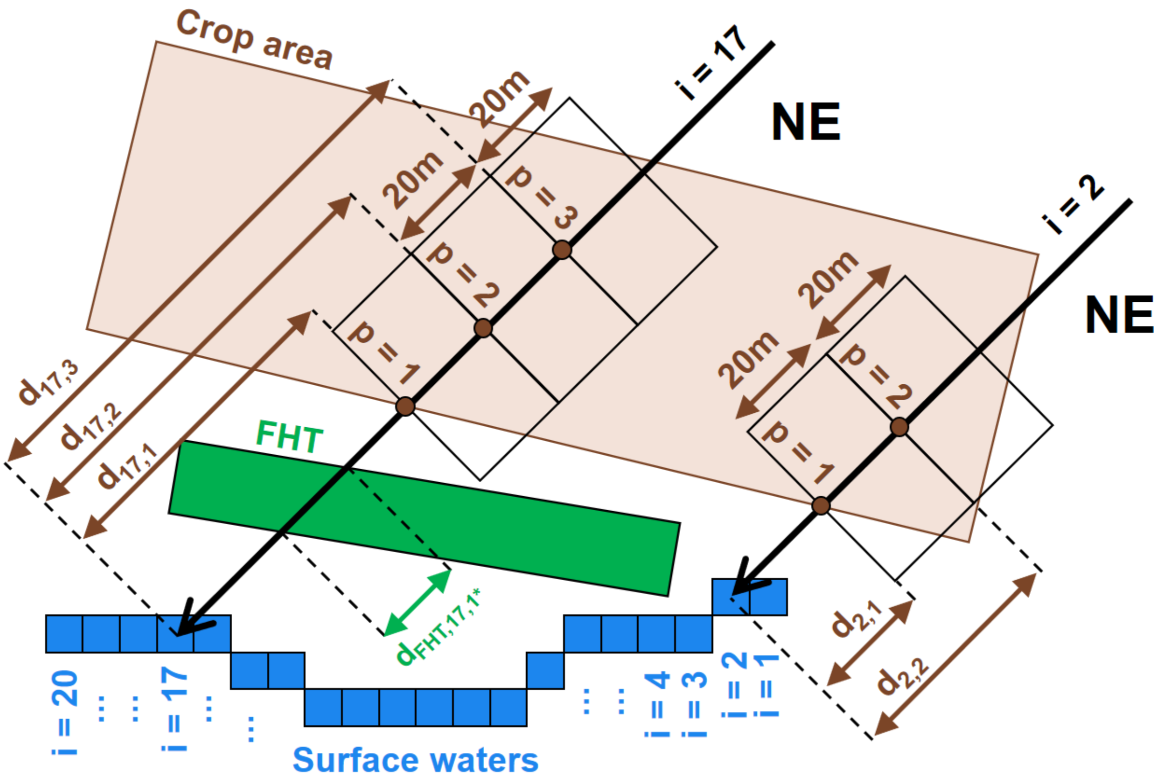


Figure 13: Example of the calculation of drift distances $d_{i,p}$ and barrier distances $d_{FHT,i,p}$ for two non-target area cells (i=2, and i=17) for the wind direction northeast (NE). In this example, the non-target area cells are surface waters. Forest, hedges and trees (FHT), but no buildings act as a barrier. *The barrier distances $d_{FHT,17,2}$ and $d_{FHT,17,3}$ are in this case equal to the barrier distance $d_{FHT,17,1}$.

3.2.2.4 Parameter selection and sensitivity analysis

The connectivity model and the spray drift model have three model parameters each. For all parameters we selected a parameter range based on field experience or literature. As a reference parameter value, we additionally selected a single value within this range that seemed the most realistic to us.

The connectivity model parameters (road carving depth d_{roads}, sink fill depth d_{sink}, and infiltration distance d_{inf}) were chosen based on our field experience from shortcut mapping and on our prior knowledge on surface runoff along roads (see Table 8). For the road carving depth $d_{roads,}$ we included the value 0 m (i.e. no change to the elevation model) as the lower end of the parameter range. However, we do not think that this value is able to represent the surface runoff accumulation effect of roads properly. To validate the results of the connectivity model, a flow path map of the study site Schalunen (Schönenberger et al., 2022a) was qualitatively compared to the model results.

In contrast to the connectivity model parameters, the spray drift model parameters were chosen based on literature values. The spray drift curves of Rautmann et al. (1999) were obtained by measuring distances up to 100 m from the sprayed plot. Therefore, for the parameter *maximal drift distance* $d_{drift,max}$, 100 m was chosen as the parameter range minimum, and as reference parameter. Since Rautmann et al. (1999) state that the curve can also be extrapolated up to 250 m, we set this distance as the parameter range maximum.

Various studies have assessed the drift intercepting properties of hedges (also known as windbreaks). For example, Wenneker et al. (2008) found a reduction of 80-90% for hedges with a width of 1 to 1.25 m in full leaf stage. Other studies report a reduction between 68% to more than 90% depending on leaf density and wind speed (Ucar and Hall, 2001). These studies show that the width of forest, hedges, or trees causing a full spray drift reduction varies depending on various factors and can therefore not be quantified by a single value. The model parameter *distance to full drift interception of forest, hedges, and trees* $d_{FHT,max}$ was therefore varied within a realistic range, based on the available data (i.e. between 5 and 20 m).

In contrast to forest, hedges, and trees, we expect buildings to completely intercept spray drift. Therefore, we set the reference parameter *drift interception by buildings* $f_{B,int}$ to 100%. However, in the sensitivity analysis, we also tested the effect of completely ignoring this process ($f_{B,int}$ = 0%).

To assess the influence of model parameters to our results, we performed a local sensitivity analysis starting from the reference parameter set and varying each parameter separately. Additionally, we combined the parameters such that they lead to extreme estimates, i.e. minimal and maximal estimate of total drift to non-target areas f_{lost} (p_{min}, p_{max}), and minimal and maximal estimate of drift deposited on surface waters relative to the drift lost on surface waters and drained roads $f_{lost,rel,SW}$ ($p_{SWrel,min}$, $p_{SWrel,max}$; see Table 8).

Table 8: Model parameters used as reference parameter set, for the sensitivity analysis, and for extreme estimates: Reference parameter set (p_{ref}), parameters used for the sensitivity analysis (p_{sens}), parameter sets for minimal and maximal total drift (p_{min}, p_{max}), parameter sets for minimal and maximal relative drift to surface waters ($p_{SWrel,min}$, $p_{SWrel,max}$). Model results were not sensitive to changes of parameters marked with a star (*) (see Sect. 3.3.3). Therefore, these parameters were kept constant when assessing the maximal and minimal drift. FHT: Forests, hedges, and trees.

Model	Parameter	p_{ref}	p_{sens}	p_{min}	p_{max}	$p_{SWrel,min}$	$p_{SWrel,max}$
Connec-tivity	Road carving depth d_{road}	10 cm	0 cm, 5 cm, 10 cm, 15 cm, 20 cm	0 cm	20 cm	20 cm	0 cm
Connec-tivity	Sink fill depth d_{sink}	20 cm	10 cm, 20 cm, 50 cm	20 cm*	20 cm*	20 cm*	20 cm*
Connec-tivity	Infiltration dist. d_{inf}	20 m	5 m, 10 m, 20 m, 30 m	10 m	30 m	30 m	10 m
Spray drift	Maximal drift distance $d_{drift,max}$	100 m	100 m, 175 m, 250 m	100 m	250 m	100 m	250 m
Spray drift	Distance of FHT causing full drift interception $d_{FHT,int}$	10 m	5 m, 10 m, 15 m, 20 m	5 m	20 m	5 m	20 m
Spray drift	Drift interception by buildings $f_{B,int}$	100%	0%, 100%	100%*	100%*	100%*	100%*

3.3 Results and discussion

3.3.1 Drainage densities

As a result of the surface runoff connectivity model, we obtained drainage densities d_s (i.e. areas of drained roads and surface waters per crop area) for each study site. The average drainage densities for arable land and vineyard sites are provided in Table 9 for the reference parameter set and the two extreme parameter sets. For all types of non-target areas, the drainage densities in vineyards are by a factor two to three higher than in arable land. This indicates that the spray drift potential in vineyard sites is higher than in arable land sites, independent of the spraying method used. Drained roads are responsible for around 73% to 84% of the total drainage density for both crop types. These results are similar to a modelling study of Alder et al. (2015), reporting that 71% of the total drainage density is caused by drained roads.

Table 9: Average drainage densities ds of arable land and vineyard sites obtained from the reference parameter set pref. In brackets, the results for the extreme parameter sets (pmin, pmax) are given.

Crop type	Roads drained to shortcuts d_{RSC}	Roads drained to surface waters d_{RSW}	All drained roads d_R	Surface waters d_{SW}	Total drainage density
Arable land	1.4% [0.81%; 2.6%]	0.11% [0.07%; 0.17%]	1.5% [0.88%; 2.8%]	0.28% [0.28%; 0.51%]	1.8% [1.2%; 3.3%]
Vineyards	4.2% [1.6%; 6.0%]	0.23% [0.28%; 0.98%]	4.4% [1.9%; 7.0%]	0.63% [0.63%; 2.6%]	5.1% [2.5%; 9.6%]

3.3.2 Spray drift losses to drained roads and surface waters

3.3.2.1 Model output example

From the spray drift model, we obtained estimates for the fraction of the applied amount lost via drift to each non-target area raster cell (either drained roads, or surface waters). In Figure 14, the spray drift model output is depicted on the example of the study site Clarmont, the wind direction southwest (SW), and the reference parameter set. The depicted part of the study site illustrates classical spray drift patterns that were also found frequently in the other study sites. Many roads are drained by storm drainage inlets (A, B) and the drift deposited per area is much higher for these roads than for surface waters (e.g. A vs. D). This can be explained by two reasons: First, drained roads are mostly situated much closer to crop areas, and they are not protected by riparian forests. Second, as mentioned in the previous section, the drainage densities are much higher for drained roads than for surface waters (Sect. 3.3.1). These factors lead to a much higher total spray drift deposition on drained roads compared to surface waters. However, this does not mean that all roads have a high potential for spray drift wash-off to surface waters. The depicted part of the study site also shows examples of drained roads receiving significantly less drift. This either is caused by larger distances between the road and the next sprayed plot along the wind line (B), by barriers that intercept spray drift (forest, hedges, trees, buildings; no example shown) or since the road is classified as undrained (C). Although undrained roads also receive spray drift from the adjacent plots, the washed off runoff is expected to infiltrate in the adjacent agricultural areas. The model results also show that depending on the wind direction, the spray drift deposition on non-target areas can vary strongly at the local scale. For example, the road areas marked with the letter B, would receive much more spray drift for the wind direction east compared to the depicted wind direction southwest.

3.3.2.2 Losses for all study sites

The modelled spray drift losses to different non-target areas are shown in Figure 15 for arable land sites, and in Figure 16 for vineyards. In vineyards, the total drift losses f_{lost} to drained roads and surface waters range between 0.063% and 0.20% on average, depending on the model parametrisation (Table 10). Almost all of these losses are deposited on drained roads. These results align well with measurements in a French vineyard catchment (Lefrancq et al., 2013) where spray drift deposition on roads amounted to between 0.07% and 0.57% of the applied amount.

Compared to vineyard sites, the average spray drift deposition to drained roads and surface waters in arable land sites is much lower, equalling between 0.0015% and 0.0049% of the applied amount. With the exception of the site Bex, all vineyard sites show larger total spray drift losses than each of the arable land sites. This difference can be explained by higher drainage densities in vineyards (see Sect. 3.3.1) and by the different application method used in vineyards (air blast sprayers instead of boom sprayers). It remains unclear, to which degree the spatial relationship between non-target areas, roads, and barriers additionally influences this result. In the study site Bex, the majority of the storm drainage system of vineyard roads drains to an infiltration basin. The density of roads drained to surface waters is therefore much smaller than in other study sites. Moreover, the closest surface waters are located far away from the vineyards. These two factors lead to very low drift losses to drained roads and surface waters in Bex.

As mentioned in Sect. 3.3.2.1, spray drift losses can vary strongly on the local scale depending on wind direction. However, our model results show that this variation is also observed at the catchment scale. These differences can amount up to a factor 4 in vineyard sites (Figure 16), and a factor 5 in arable land sites (Figure 15). On average, the difference between the wind direction with the highest and lowest spray drift deposition equals a factor of 2.2. For certain study sites, spray drift deposition on drained roads and surface waters could therefore be reduced considerably by applying pesticides during favourable wind directions.

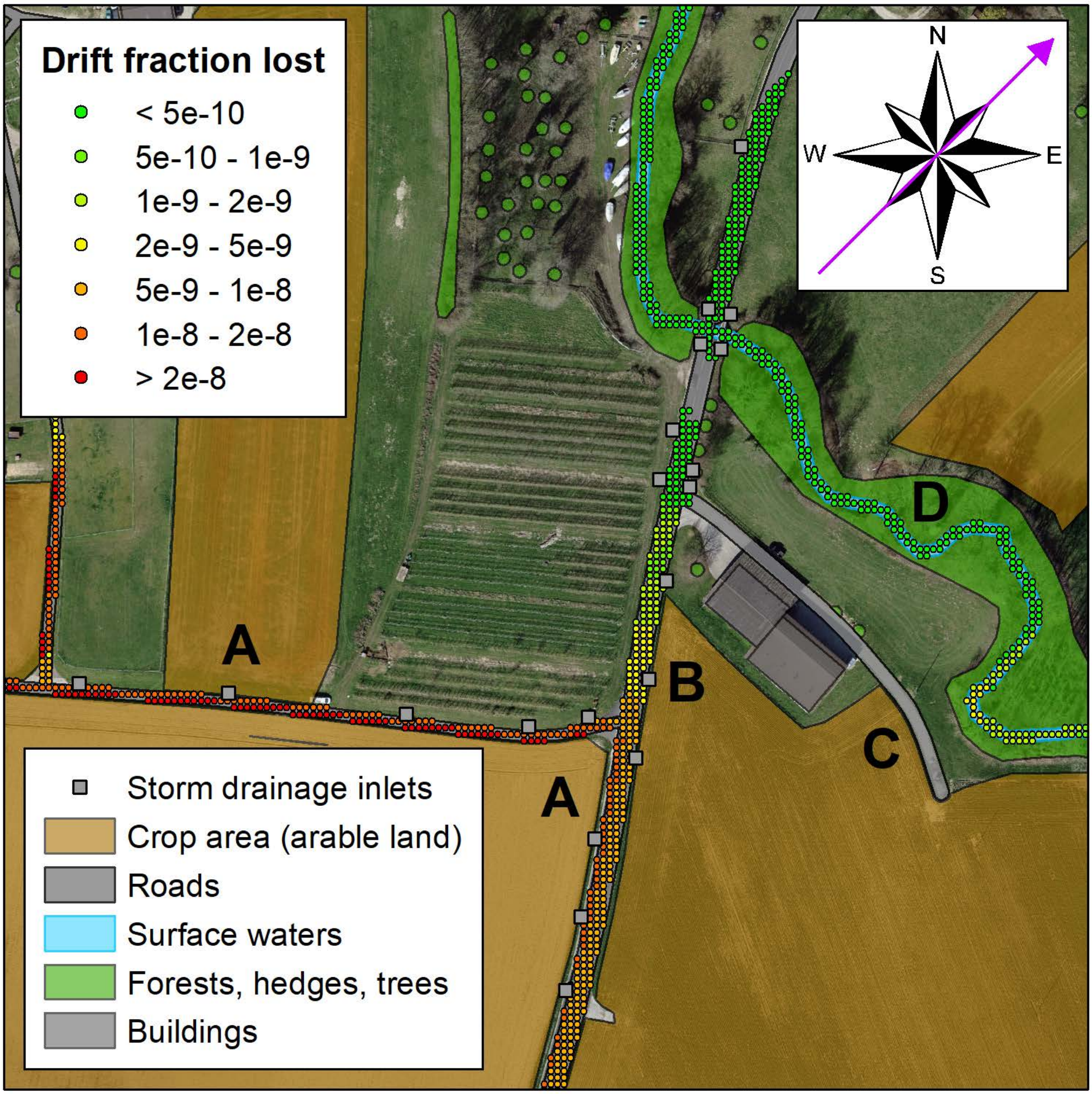


Figure 14: Spray drift model results for the reference parameter set and the wind direction southwest, for a part of the study site Clarmont. The values reported represent the fraction of drift lost to each non-target area cell relative to the total amount applied in the whole study site. Sources: Kanton Aargau et al. (2020); Swisstopo (2019b); Swisstopo (2020b)

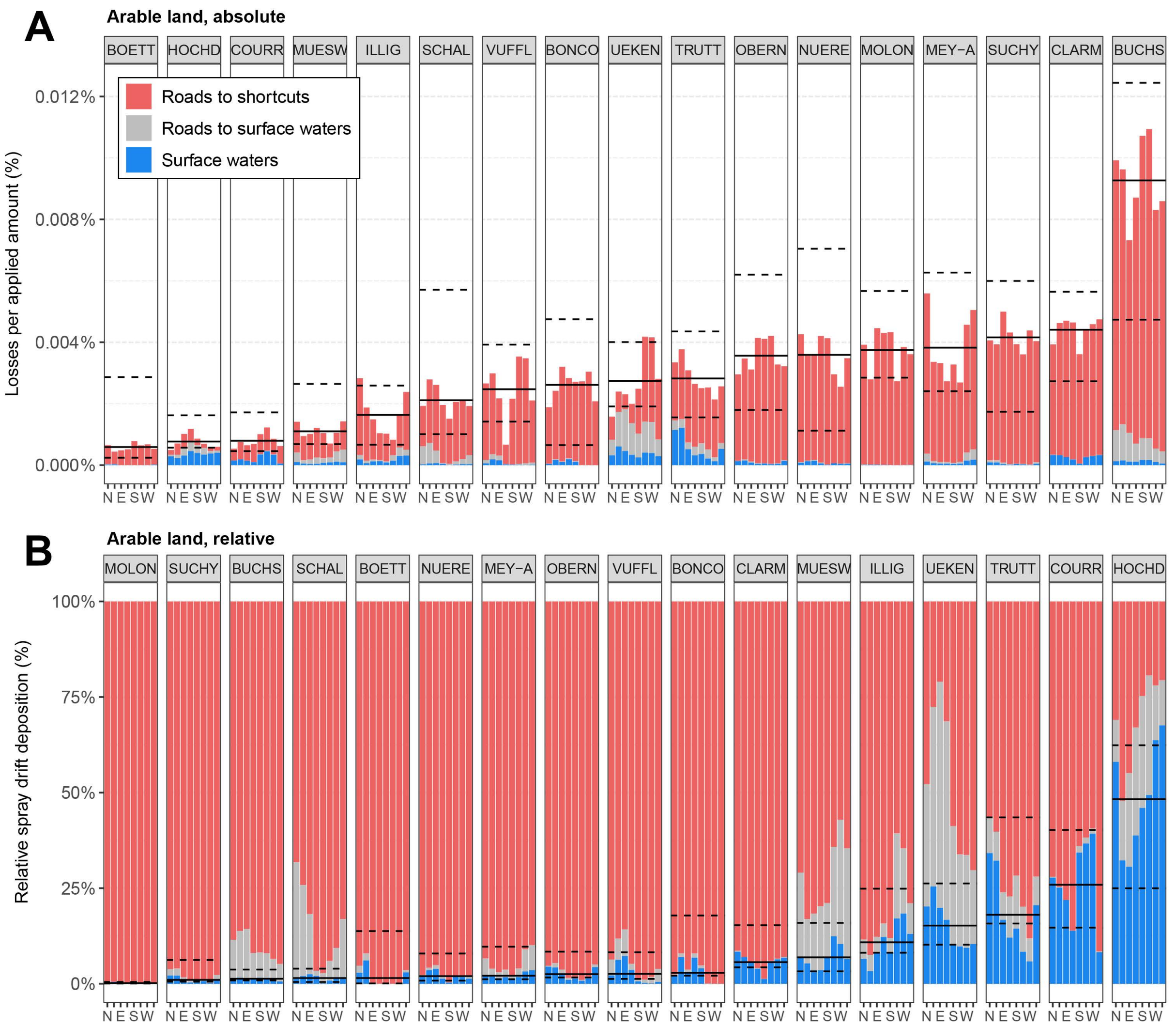


Figure 15: Drift losses in arable land study sites per wind direction: (A) Fraction lost per total amount applied on arable crops ($f_{lost,w}$). The black solid lines indicate the total losses resulting from the reference parameter set (p_{ref}). The dashed lines report the losses from the extreme parameter sets (p_{min}, p_{max}). (B) Losses per non-target area, relative to the losses to all three non-target areas ($f_{lost,rel,w}$). The black solid lines indicate the losses to surface waters, resulting from the reference parameter set (p_{ref}). The dashed lines represent the losses to surface waters resulting from the extreme parameter sets ($p_{SWrel,min}$, $p_{SWrel,max}$).

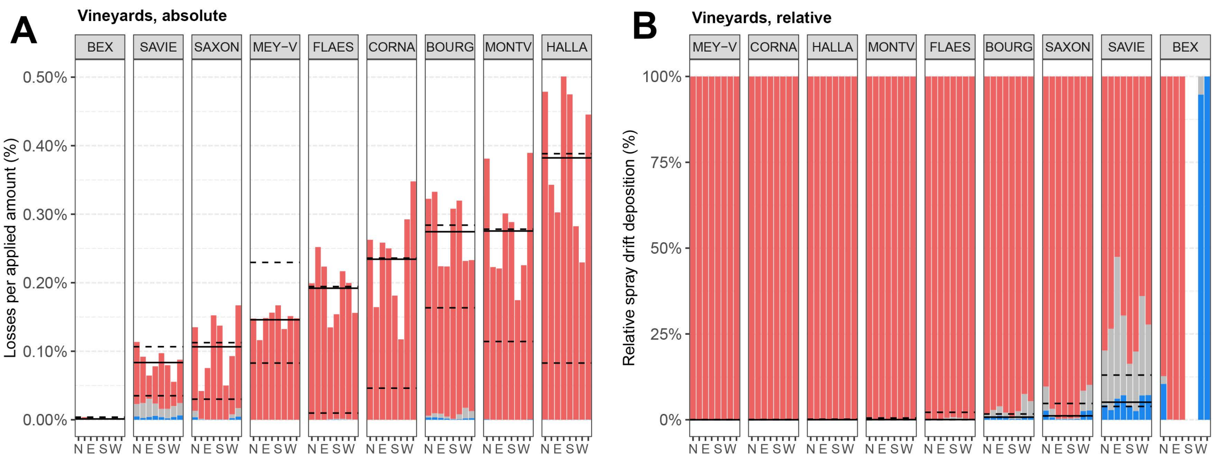


Figure 16: Drift losses in vineyard study sites per each wind direction: (A) Fraction lost per total amount applied in vineyards ($f_{lost,w}$). The black solid lines indicate the total losses resulting from the reference parameter set (p_{ref}). The dashed lines report the losses from the extreme parameter sets (p_{min}, p_{max}). (B) Losses per non-target area, relative to the losses to all three non-target areas ($f_{lost,rel,w}$). The black solid lines indicate the losses to surface waters, resulting from the reference parameter set (p_{ref}). The dashed lines represent the losses to surface waters resulting from the extreme parameter sets ($p_{SWrel,min}$, $p_{SWrel,max}$).

A comparison of the relative spray drift losses to drained roads and to surface waters reveals that most of the spray drift is deposited on drained roads (average – 82 to 95% for arable land, 97 to 99% for vineyards; Table 10). Accordingly, the spray drift deposited on drained roads that potentially can be washed off to surface waters is much larger than the spray drift directly deposited in surface waters for both crop types, provided that the legally required buffer distances are kept (see discussion in next paragraph). As shown in Figure 15 and Figure 16, most of the spray drift deposition on drained roads is taking place on roads drained by shortcuts. Shortcuts therefore strongly increase the potential of spray drift wash-off from roads to surface waters.

In our analysis, we assumed that farmers comply with the buffer distances according to Swiss regulations and to Swiss proof of ecological performance (see Sect. 3.2.2.3). However, these buffer distances to surface waters are often not kept in Swiss vineyards. Therefore, the above-mentioned results represent an ideal situation for the drift to surface waters and the real drift to surface waters is higher.

In the timespan between the application and the next rain event, different degradation processes (e.g. photolysis or transformation related to concrete alkalinity), but also sorption may lead to a significant reduction of the spray drift available for wash-off from roads (Jiang and Gan, 2016). From the drift losses to drained roads, the majority (75%) is deposited on asphalt or concrete roads. During experiments on such roads, 57% or more of the amount applied of substances with low soil adsorption coefficients ($K_{oc} < 250$ mL g^{-1}) was washed off (Ramwell et al., 2002; Thuyet et al., 2012). In these studies, the time between application and rainfall amounted between six hours and seven days, being a realistic range for the time elapsing between application and rainfall in many parts of Western and Central Europe. For substances with low K_{oc}, we therefore expect the amount of spray drift washed off from drained roads to clearly exceed the amount of spray drift directly deposited in streams. In contrast, for substances with higher K_{oc}, maximal wash-off reported during the first rainfall events after application reached up to 5.8% (Thuyet et al., 2012) and 2.7% (Jiang et al., 2012). For these substances, we therefore expect the amount of spray drift washed off from roads to surface waters to be in the same order of magnitude or lower than the direct spray drift to surface waters. However, it should be kept in mind that such substances might still be washed off during later rain events with the road acting as a pesticide reservoir (Jiang and Gan, 2016).

To determine the relevance of spray drift wash-off from drained roads for the total pesticide load in the stream, we compared the spray drift losses to total loss rates to surface waters. Total losses to surface waters typically range between 0.005% and 1% of the applied amount (Doppler et al., 2014; Leu et al., 2004a; Riise et al., 2004; Siimes et al., 2006). Therefore, in arable land sites, the spray drift losses to drained roads (0.0015% and 0.0049%) are small compared to typical total loss rates. However, in vineyard sites, the losses to drained roads (0.063% to 0.20%) represent a major fraction compared to typical total loss rates. For certain substances (see above), we therefore expect the wash-off from drained roads to be a relevant transport pathway compared to total pesticide losses to surface waters. Furthermore, it is important to note that between two rain events spray drift losses to drained roads are accumulating and are then all washed off at once. This might lead to much higher concentration peaks than direct spray drift deposition to surface waters during single spray applications.

Previous studies have shown that spray drift to roads can be an important transport pathway in single catchments (Ammann et al., 2020; Lefrancq et al., 2014; Rübel, 1999). Our results indicate that for catchments with high densities of drained roads and for application methods with a high spray drift potential, these findings can be generalized.

Table 10: Average drift to drained roads and surface waters for arable land and vineyard sites. The reported values indicate the results of the reference parameter set. In brackets, the results of the extreme parameter sets are given. For the calculation of relative losses in vineyard sites, the study site Bex was excluded.

Target area	All drained roads		Surface waters	
	Losses per applied amount $f_{lost,R}$ (-)	Relative loss on non-target area type $f_{lost,rel,RSC} + f_{lost,rel,RSW}$	Losses per applied amount $f_{lost,SW}$ (-)	Relative loss on non-target area type $f_{lost,rel,SW}$
Arable land	$2.8 \cdot 10^{-5}$ [$1.4 \cdot 10^{-5}$; $4.6 \cdot 10^{-5}$]	91.3% [81.8%; 94.7%]	$1.5 \cdot 10^{-6}$ [$1.3 \cdot 10^{-6}$; $2.9 \cdot 10^{-6}$]	8.7% [5.3%; 18.2%]
Vineyards	$1.8 \cdot 10^{-3}$ [$6.2 \cdot 10^{-4}$; $2.0 \cdot 10^{-3}$]	99.1% [97.2%; 99.3%]	$9.0 \cdot 10^{-6}$ [$8.7 \cdot 10^{-6}$; $1.2 \cdot 10^{-5}$]	0.9% [0.7%; 2.8%]

3.3.3 Model uncertainties

In the previous section, model uncertainty was addressed by reporting the results as a range between the minimal and maximal parameter sets (p_{min}, p_{max}, $p_{SWrel,min}$, $p_{SWrel,max}$). In the following, we elaborate on the importance of single model parameters on the overall uncertainty and on additional uncertainties related to the models used in this study.

The combined sensitivity analysis of the surface runoff connectivity and the spray drift model shows that the parameters road carving depth d_{road} and infiltration distance d_{inf} cause the largest model uncertainties (details – Sect. S3.2). These two parameters are both used to classify roads as drained or undrained in the surface runoff connectivity model. This indicates that the classification of roads is one of the major uncertainty factors. To check the plausibility of road classification, we therefore compared the areas classified as drained roads to flow paths mapped during a snowmelt event on 12 March 2018 in the study site Schalunen (Schönenberger et al., 2022a). This comparison suggests that the road areas drained by shortcuts are underestimated by the reference parameter set and that they are rather in the range of the values resulting from the maximal total drift parameter set (p_{max}). However, during this snowmelt event, the amount of runoff on roads was exceptionally high. Accordingly, we expect that flow paths were longer during the snowmelt event than during most rain events. Therefore, this comparison affirms the plausibility of the range of model outputs. Nevertheless, the classification of roads as drained or undrained remains a major source of uncertainty. Further studies on spray drift wash-off from roads should therefore address this issue, for example by extensive mapping of flow paths during rain events.

Additional uncertainties are caused by the extrapolation of the spray drift curves to a minimal drift distance of 0.5 m. During the field trials used for spray drift curve determination, the minimal drift distances measured were 1 m (arable land) and 3 m (vineyards) from the sprayed plot. Since the buffer width around surface waters equals 6 m, this extrapolation was only used for estimating spray drift to drained roads, but not to surface waters. If the effective drift curve is below the extrapolated drift curve (eq. 3.2) for distances shorter than the minimal measured distances, our model would lead to an overestimation of the spray drift to drained roads. To ensure that our conclusions are not an artefact of the spray drift curve extrapolation, we performed another model run using the reference parameter set. However, for distances smaller than the minimal measured distances, we did not use the extrapolated spray drift curve, but restricted the spray drift deposition to the values at the minimal measured distance (1 m/3 m). For arable land sites, this led to a reduction of only 2.5% of the estimated drift losses to drained roads. The extrapolation uncertainty can therefore be neglected for this crop type. However, for vineyards, a much larger extrapolation uncertainty (reduction of 51%) was found. This uncertainty is not large enough to change the conclusions drawn on the potential of spray drift wash-off from drained roads in vineyards (Sect. 3.3.2.2). However, to reduce the uncertainty in the estimation of spray drift deposition on vineyard roads, additional drift trials in ultimate proximity of vineyard plots (< 3 m) would be needed.

In this study, we assumed that pesticides are not applied within a buffer of 6 m around surface waters. However, several pesticides are only authorized for usage outside of larger buffers (20 m, 50 m, or 100 m). For these pesticides, the direct drift to surface waters is much lower and the relative importance of drift wash-off from roads is much higher. This further underlines the high potential of spray drift wash-off from roads compared to direct drift to surface waters.

3.3.4 Implications for practice

The results presented in this study suggest that spray drift wash-off from drained roads is a major source for the pesticide pollution of surface waters, at least in vineyards and for pesticides with low K_{oc}. To reduce spray drift to drained roads, various measures could be worth considering. These measures include drift reducing spraying techniques, and drift barriers or buffer strips between the sprayed plots and drained roads. We used the spray drift model presented here to assess the potential of buffer strips for reducing spray drift to drained roads. For the reference parameter set, our model predicts that a 3 m buffer around all drained roads would lead to a 37% and 74% reduction of spray drift in arable land, and vineyard sites, respectively. With a 6 m buffer, spray drift to drained roads would be reduced by 56% and 90%. However, it has to be kept in mind that especially for vineyards, the spray drift curves are rather uncertain for distances smaller than 3 m (see Sect. 3.3.3).

Spray drift to drained roads and subsequent wash-off is currently not considered in European or Swiss pesticide authorisation and legislation. Our results however indicate that this transport pathway is relevant, at least in certain cases. This demonstrates that current regulations only cover a part of the total pesticide transport to surface waters related to spray drift. The same issue has been shown for the surface runoff related transport of pesticides via shortcuts (Schönenberger et al., 2022a; Schönenberger and Stamm, 2021). Authorities should therefore consider the potential of pesticide transport via shortcuts in the pesticide registration process and when designing regulations. At the same time, farmers should be aware of the potential of this process when applying pesticides.

3.4 Conclusions

- In agricultural catchments in Switzerland, many roads are drained by shortcuts (storm drainage system inlets, channel drains, ditches) or directly to surface waters. The density of such roads is 2.7 to 7 times larger than the density of surface waters.
- The amount of spray drift deposited on drained roads is much larger than the direct drift deposition in surface waters. In the arable land sites studied, spray drift to drained roads exceeded the direct drift by a factor of 4.5 to 18. In vineyard sites, this factor amounts between 35 and 140, assuming that farmers comply with the legally required buffer distances. Most spray drift losses to drained roads are deposited on roads drained by shortcuts, and only a minor part is deposited on roads directly draining to surface waters.
- Compared to typical total pesticide loss rates to surface waters, the spray drift losses to drained roads are rather small in arable land sites (losses equal between 0.0015% and 0.0049% of the applied amount). However, in vineyard sites, the losses to drained roads are substantial (0.063% to 0.20% of the applied amount).
- Current literature suggests that major fractions of the spray drift on roads can be washed off during subsequent rain events, especially for pesticides with low soil adsorption coefficients (K_{oc}). Especially for such pesticides and in vineyards, the spray drift wash-off from drained roads is therefore expected to be a relevant transport pathway to surface waters.
- These findings should be considered for adapting pesticide registration procedures and for implementing best management practices in critical agricultural areas such as vineyards.

Chapter 4. Pesticide concentrations in agricultural storm drainage inlets of a small Swiss catchment

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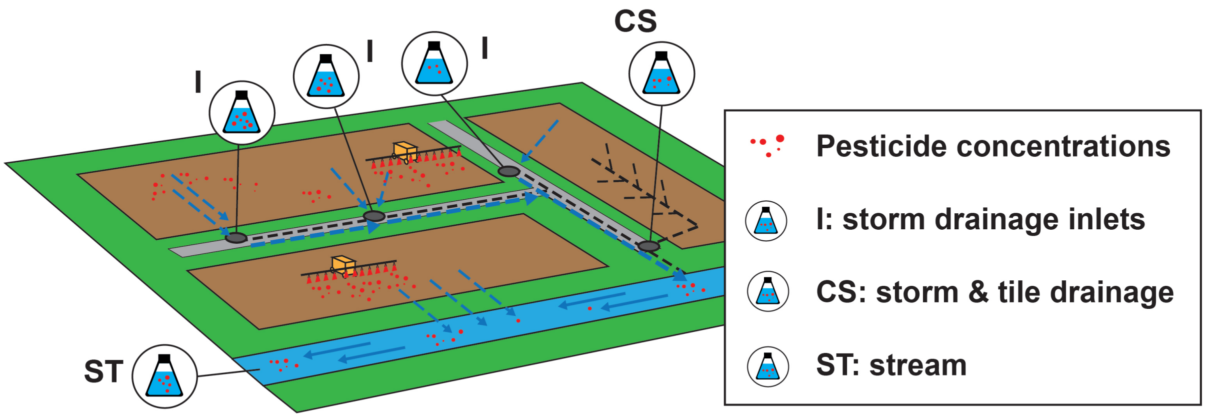
Graphical abstract



Abstract

Agricultural pesticides transported to surface waters pose a major risk for aquatic ecosystems. Modelling studies indicate that the inlets of agricultural storm drainage systems can considerably increase the connectivity of surface runoff and pesticides to surface waters. These model results have however not yet been validated with field measurements. In this study, we measured discharge and concentrations of 51 pesticides in four out of 158 storm drainage inlets of a small Swiss agricultural catchment (2.8 km^2) and in the receiving stream. For this, we performed an event-triggered sampling during 19 rain events and collected plot-specific pesticide application data. Our results show that agricultural storm drainage inlets strongly influence surface runoff and pesticide transport in the study catchment. The concentrations of single pesticides in inlets amounted up to 62 µg/L. During some rain events, transport through single inlets caused more than 10% of the stream load of certain pesticides. An extrapolation to the entire catchment suggests that during selected events on average 30% to 70% of the load in the stream was transported through inlets. Pesticide applications on fields with surface runoff or spray drift potential to inlets led to increased concentrations in the corresponding inlets. Overall, this study corroborates the relevance of such inlets for pesticide transport by establishing a connectivity between fields and surface waters, and by their potential to deliver substantial pesticide loads to surface waters.

4.1 Introduction

Pesticides used in agriculture impair water quality, leading to biodiversity losses in aquatic ecosystems and threaten drinking water resources (Kiefer et al., 2020; Sánchez-Bayo and Wyckhuys, 2019; Stehle and Schulz, 2015). To protect surface waters from those negative impacts with appropriate measures, it is essential to understand how pesticides are transported from the field to surface waters. Current research usually distinguishes between two types of pesticide transport pathways: Point sources and diffuse sources. Farmyard runoff (De Wilde et al., 2007; Reichenberger et al., 2007), wastewater treatment plants (Eggen et al., 2014; Munz et al., 2017), combined sewer overflows (Mutzner et al., 2020; Neumann et al., 2002) or accidental spills (Reichenberger et al., 2007) are considered as the most important point sources. For diffuse sources, surface runoff (Larsbo et al., 2016; Lefrancq et al., 2017), spray drift (Lefrancq et al., 2013; Vischetti et al., 2008), and macropore flow to tile drainages (Sandin et al., 2018) are considered of major importance.

Pesticide transport from diffuse sources has been shown to be strongly influenced by artificial structures affecting the connectivity between fields and the stream network (Frey et al., 2009). For example, in several studies, roads and ditches were shown to concentrate surface runoff and increase pesticide losses (Fiener et al., 2011; Heathwaite et al., 2005; Hösl et al., 2012; Payraudeau et al., 2009; Rübel, 1999). Additionally, in a French vineyard, spray drift on roads and subsequent wash off was found to be a major pesticide transport pathway (Lefrancq et al., 2014). In contrast to other countries, roads and adjacent fields in Switzerland are less often drained to ditches, but to inlet and maintenance shafts of storm and tile drainage systems (Alder et al., 2015). In a model-based study on the national level, we found that around half of surface runoff from fields and the related pesticide load is expected to be transported to surface waters through such shafts (Schönenberger and Stamm, 2021). Similarly, another model-based study suggests that also the wash-off of spray drift deposited on roads through such shafts to surface waters may be a major pesticide transport pathway (Schönenberger et al., 2022b). However, there is a lack of empirical data to validate these findings. So far, field data on transport of agricultural pollutants through inlet or maintenance shafts were only reported in two studies. Firstly, Remund et al. (2021) performed a long-term study on soil erosion in five Swiss study catchments. They found that 88% of the sediment and phosphorus losses from arable land to surface waters occurred through inlet or maintenance shafts. Secondly, Doppler et al. (2012) measured pesticide concentrations in the stream and the underground pipe system of a small Swiss agricultural catchment. They found that inlet shafts, maintenance shafts and the connected pipe system were creating shortcuts between remote areas of the catchment and the stream, enabling fast transport of surface runoff and pesticides. Inlet and maintenance shafts were therefore called hydraulic shortcuts.

Although the above-mentioned studies indicate that hydraulic shortcuts can be a relevant transport pathway, direct measurements of surface runoff and pesticides transported through hydraulic shortcuts in agricultural areas currently do not exist. To close this gap, we measured runoff and pesticide transport through inlet shafts (or simply inlets in the following, see Figure 17A) of an agricultural storm drainage system for the first time. The measurements were performed in a catchment in which we expected rather high pesticide transport through hydraulic shortcuts (i.e. an intensively used agricultural catchment with a high shortcut density). We focussed our study on inlets, since this type of hydraulic shortcut was identified as the most important shortcut type in a previous study (Schönenberger and Stamm, 2021).

Therefore, we aimed on answering the following research questions:

1) How often is surface runoff transported through storm drainage inlets and which ratio of the discharge in the stream is caused by this process?
2) Which pesticide concentrations and loads are transported during selected rain events?
3) How are transport pathways, pesticide applications, and substance properties affecting pesticide concentrations in inlets?

To answer these questions, we focused on a study catchment with a high number of shortcuts and little direct surface connectivity to the stream. However, the conditions in the study catchment (soils, topography, climate, storm drainage system) are quite typical for the Swiss Plateau such that key findings can be generalized to a larger area.

4.2 Material and Methods

4.2.1 Study catchment

The study catchment (Figure 18) is located in a rural area in the Swiss midlands (canton of Bern, outlet: 47°07′12.570″N 7°30′48.926″E). It has a size of 2.8 km^2 and is covered by arable land (38%), forests (32%), agricultural areas with very little or no pesticide use (18%) (e.g. meadows, pasture, ecological compensation areas), and other/undefined agricultural areas (4%). Settlements, farmyards, roads and farm tracks mainly cover the remaining area (8%). On arable land, the predominant crop types during the study year were grains, potatoes, and sugar beets. The average annual rainfall equals 1075 ± 163 mm/yr (MeteoSwiss, 2018) and the average slope is 5.0%. The agricultural area is heavily drained by artificial structures by tile drains in the soils and by storm drains along the road network. In total, 158 storm drainage inlets (see Figure 17A) were identified along or on agricultural areas.. Most of them are located along farm tracks (111), or concrete roads (33). The remaining fourteen are located directly on fields. All of these inlets are drained to the stream at the catchment outlet. In addition, 84% of the agricultural area is tile drained.

Most of the 26 farmers in the catchment were participating in a program aiming to reduce pesticide pollution in the receiving stream. They had the freedom to decide on pesticide applications themselves, but received subsidies for reduction measures (e.g. creating buffer strips or reducing herbicide use). We received plot-specific crop and pesticide application data for 96% of the agricultural areas in the catchment for the period January to October of the study year 2019. The pesticide application data was recorded by the farmers using a crop management system and included the day of application, product, amount applied, crop, plot size, and a georeferenced polygon of the plot.

4.2.2 Field work

4.2.2.1 Sampling site selection

We selected six sampling sites in the catchment (see Figure 17B and Figure 18). Four were located at storm drainage inlets (I1-I4), and one each at a collector shaft (CS) and the stream (ST) at the catchment outlet.

I1-I4 were selected as follows from the 158 inlets in the catchment. To be a suitable sampling location, an inlet had to fulfil two criteria. First, the dimensions of the inlet had to allow the installation of measuring equipment. Second, we aimed on sampling only surface runoff entering the inlet through the lid, but no other inflows. To ensure that no tile drainage flow enters the inlet, we therefore also excluded all inlets with inflow pipes. From the ten inlets fulfilling these criteria, we selected the four that represented the different terrain and cropping conditions best (see Figure S48 to Figure S51). They are all located at the border of a field and a gravel farm track. While I1, I2, and I4 are lying directly next to the farm track, I3 is separated from the farm track by a grass strip of approximately 0.5 m width (Figure S50). During dry periods, there is no discharge transported through the four inlets, and in I1, I2, and I4 the water stagnates at the height of the outlet pipe (Figure S52). In contrast, during dry periods, the water level in I3 falls to a lower level due to seepage through the shaft bottom.

Because of the second selection criterion, the selected inlets only cover a small fraction of the total surface runoff transported through storm drainage inlets in the catchment. By measuring in shafts collecting storm drainage water from several inlets, we could have increased the fraction of surface runoff sampled. However, in most shafts it was not possible to distinguish if an inflow pipe is only connected to storm drainage inlets, or also to the tile drainage system. The restriction of our measurements to inlets without inflow pipes was therefore necessary to ensure that our signal only consists of surface runoff.

4.2.2.2 Installations

Inlets (I1-I4): In each inlet, we measured discharge by installing a weir with a calibrated rating curve in front of the outlet pipe. The water level was measured using a capacitive pressure sensor (DWL compact, UIT, Germany) coupled to a data logger equipped with a GPRS module (LogTrans-field, UIT, Germany). For water sampling, we installed an event-based, water-level proportional sampler (details – Sect. S4.1.1.2). The GPRS module was used for triggering other samplers (details – Sect. 4.2.2.3), data transfer, and to inform scientists.

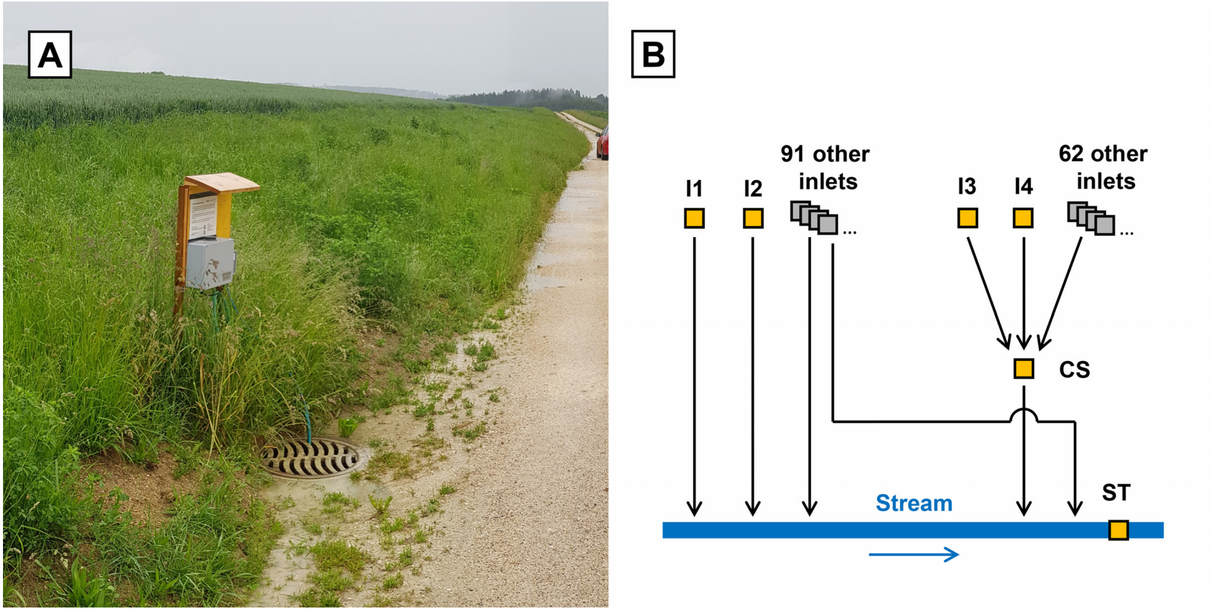


Figure 17: (A) Example picture of a storm drainage inlet in the study catchment taken during the study period. The depicted inlet (I1) is one of the four inlets sampled and is situated between a farm track and a wheat field. A larger picture of the situation around the inlet is shown in Figure S48. (B) Schematic representation of the storm drainage network in the catchment (black lines: pipes, grey squares: inlets) and of the sampling locations (yellow squares). I1-I4: inlets, CS: collector shaft, ST: stream.

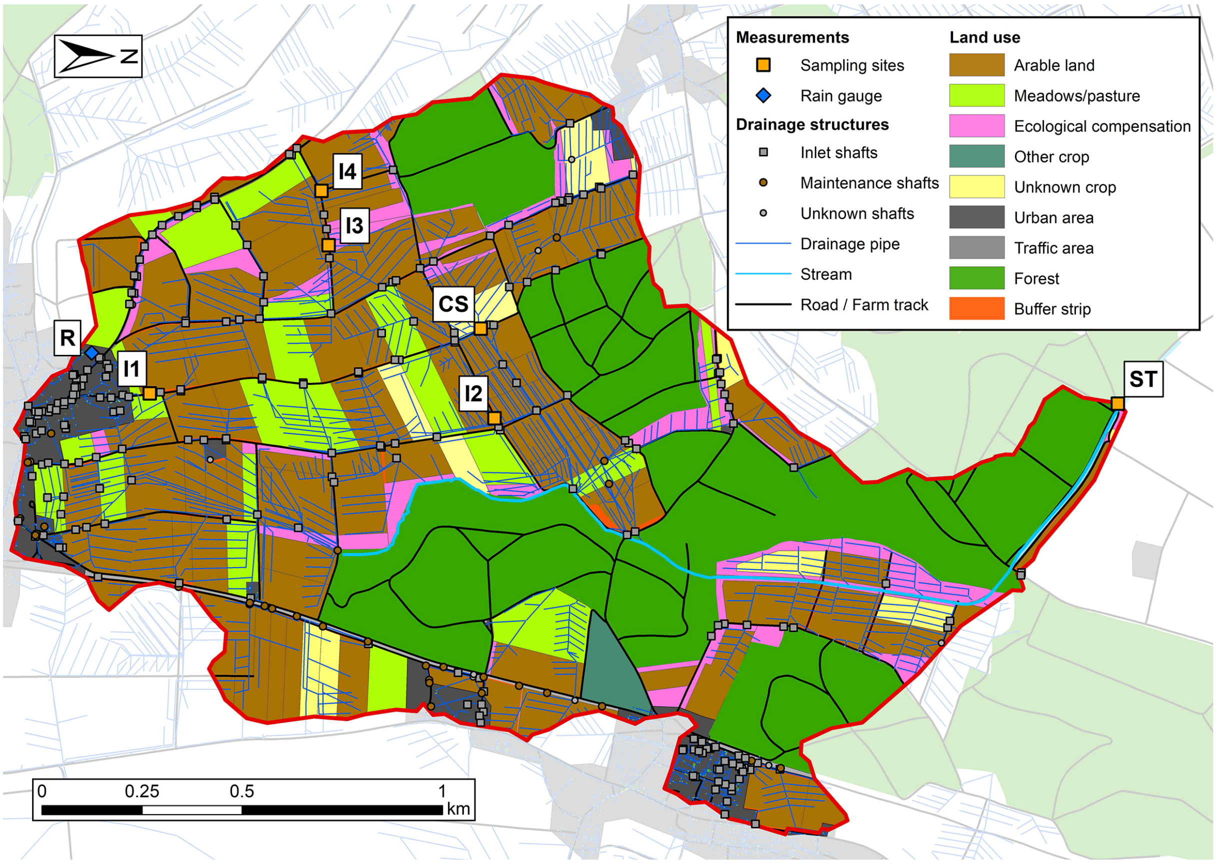


Figure 18: Map of the study catchment. Yellow squares show the sampling sites (I1-I4: inlets, CS: collector shaft, ST: stream) and the blue diamond shows the rain gauge (R). Source of background map: Swisstopo (2020b).

Collector shaft (CS): This shaft collects water from 64 inlets (including I3 and I4), and from a large part of the tile drainage system in the catchment (Figure S53 and Figure S54). At this site, the water level was measured using the same sensors as in the inlets. Water samples were taken using an automatic sampler (TP5C portable sampler, MAXX GmbH, Germany) coupled to a GPRS module.

Stream (ST): At the catchment outlet (Figure S55), discharge was measured by the cantonal authorities using an ultrasonic sensor (POA-V2XXK, NIVUS AG, Germany). Water samples were taken with the same sampler type as in the collector shaft.

Rain gauge (R): Rainfall data (resolution: 1 min, accuracy: 0.1 mm) was provided by the cantonal authorities from a rain gauge at the southern catchment edge.

4.2.2.3 Sampling strategy

In central Europe, most pesticides are applied in spring and summer (Halbach et al., 2021; Szöcs et al., 2017) and rainfall intensities are higher during this time of the year (Umbricht et al., 2013). Consequently, the highest pesticide concentrations in surface waters are usually measured during this period (Doppler et al., 2017). We therefore selected our study period (1 April to 20 August 2019) such that it covers most of this high-risk period. From the substances analysed in this study, 96% of the total active ingredient mass applied in 2019 was applied within this period (see Figure S58). Since water only flows through the inlets during rain events, we performed an event-based sampling.

In the inlets, the water-level proportional samplers started sampling at a defined water level threshold above the bottom of the weir (2 cm for inlets with little runoff, 3 cm for inlets with larger runoff), corresponding to a discharge of approximately 1.7 and 5 L/min. This resulted in one composite sample per event for each inlet exceeding the water level threshold. Rain events that were too small to exceed the water level threshold in an inlet were not sampled. When the water level threshold was exceeded in at least two inlets, the automatic samplers at the collector shaft and the stream were triggered via the GPRS module to start sampling (see Figure S57). In the collector shaft, time proportional samples (50 mL) were taken every 2 to 3 minutes and pooled together into one composite sample per 20 to 30 minutes, depending on the event (details – Table S16). Depending on the event duration, the total sampling duration was 4 to 8 hours. In the stream, time proportional sampling was performed with the same frequency during the discharge peak. Before and after the peak, samples were pooled over a period of up to two hours. Depending on the event duration, the total sampling duration was 10 to 12 hours. All samples were kept in glass bottles and protected from sunlight. At sites CS and ST, the samples were cooled by the automatic samplers (4°C), and at sites I1-I4 by the stagnating water around the bottle (average temperature: 13.5°C). They were collected on average 1.3 days after sampling and frozen at -20°C until analysis. If no composite samples were taken in an inlet during an event (due to lack of sufficient discharge, or due to malfunctioning of the sampler), we took a grab sample from the stagnant water during sample collection. Cantonal authorities were also taking samples in the stream (15 min sampling interval, eight hour composite samples) serving as a backup in case of malfunctioning of the automatic sampler.

4.2.2.4 Field mapping

During a snowmelt event on 12 March 2018, we mapped the surface runoff pathways in a part of the catchment (Figure S59). We chose a snowmelt event for this mapping campaign, since it was easier to predict snowmelt events than intense rainfall events generating surface runoff. Since runoff pathways strongly depend on the amount of runoff formed, this mapping campaign only provides a rough estimation of the potential runoff pathways during rain events.

4.2.3 Chemical analysis

Overall, we collected 423 samples and selected 193 of them as the most relevant ones (see below) for further analysis. Most importantly, we analysed all inlet samples. In a second step, we analysed collector shaft and stream samples for six out of the top ten events with the highest sum concentrations in the sampled inlets, such that they cover the range of rain intensities observed (details Table S16). For the selected samples, dissolved phase pesticide concentrations were determined using direct injection liquid chromatography coupled to high-resolution mass spectrometry (LC-HRMS). The particulate phase was not analysed. The target list (Table S13) included 51 substances that were either pesticides known to be applied in the catchment (45 substances) or their transformation products (6 substances). Samples were thawed and centrifuged for 5 minutes at 2000 g. The supernatant was transferred and isotope-labelled internal standard (ISTD) was spiked (details – Table S13). Randomly selected samples were spiked with a standard solution in order to assess relative recovery of the compounds. Centrifugation, transfer, spiking of ISTD and standard solution were performed by a fully automated workflow. Laboratory blanks and blinds, and field blinds were included in the measurement sequence to monitor instrument carry-over and contamination. Chromatographic separation was performed on a reversed-phase C18 column (Atlantis T3, 3 µm particle size, 3.0x150 mm inner diameter, Waters), applying a water-methanol gradient (both containing 0.1% formic acid). The measurements were performed on a hybrid quadrupole-orbitrap mass spectrometer (Lumos Fusion, Thermo Scientific) equipped with an electrospray ionisation source. Quantification of the target compounds was performed using TraceFinder 5.1 (Thermo Scientific). For 95% of the compounds, relative recovery was in the range of 80-120%. For 80% of the compounds, the limit of quantification (LOQ) was 20 ng/L or lower. Further details on the chemical analysis (such as the gradient, the ionization, processed sample volumes) are given in Sect. S4.1.2

4.2.4 Data analysis

4.2.4.1 Surface runoff connectivity

To determine the topographical catchment of each sampling site with respective crops and pesticide applications, we used a surface runoff connectivity model (Schönenberger and Stamm, 2021). The model is based on a digital elevation model (Swisstopo, 2019a) with 2 x 2 m resolution and a D-infinity flow algorithm (Tarboton, 1997). Despite the high spatial resolution, it cannot represent all microtopographical features such as subtle depressions or the effects of roads. These sub-grid effects are represented by average effects in the model parameterisation. We adjusted the model parameters (e.g. road carving depth, or sink filling depth) such that the output fitted the observed flow paths in the field well (detail – Table S11).

The model output indicates from which agricultural areas (called contributing areas in the following) surface runoff drains to a particular inlet or directly to the stream, and from which areas surface runoff infiltrates in a sink. We intersected the contributing areas with the plot-specific crop and pesticide application data. This provided us with an estimate of crops planted and pesticides applied in the contributing area of each inlet, sink, and the stream.

In addition, we performed a Monte Carlo simulation of the surface runoff connectivity model with 100 model runs. The parametrization was identical as in Schönenberger and Stamm (2021). This allowed us to assess the uncertainty introduced by the model parameter selection, and to compare the connectivity in the study catchment to the national assessment of the mentioned study.

4.2.4.2 Definition of events

We classified two types of events – rainfall and sampling events. Measured rainfall was classified into a rainfall event if the total rainfall exceeded 1 mm within 8 hours. Subsequent rainfall was assigned to the same event if there was no dry period of at least 8 hours in between. After dry periods of more than 8 hours, a new rainfall event was defined. Sampling events were defined as rainfall events during which water samples were taken.

4.2.4.3 Transport processes

For each measured pesticide in a sample, we determined potential transport processes causing the measured concentration. Based on the spatio-temporal relation between samples and applications, we assigned each concentration measurement to one of the following categories: A) No reported application, B) other, C) spray drift / other, D) surface runoff / (tile drainage) / spray drift / other. In the following, we explain these categories and how they were assigned.

A) *No reported application*. If the pesticide was not applied in the catchment during the study year, or only after the sample was taken, the measured concentration was assigned to this category. Concentrations in this category may be due to wash-off of residuals from previous year's applications, originate from unreported applications, or may relate to applications outside the study catchment (e.g. atmospheric deposition).

B) *Other*. This category was assigned if the pesticide was applied in the catchment before the sampled event, but on a field not allowing for transport via spray drift, surface runoff or tile drainages to the sampling site. Concentrations in this category may originate from droplet losses from leaky spraying equipment, farmyard runoff, accidental spills, atmospheric deposition, or a process mentioned in the previous category.

C) *Spray drift / other*. This category was assigned if the pesticide was applied before the event and spray drift to the sampling site was possible, but not transport via surface runoff or tile drainages. In the study catchment, only ground applications are performed and spray drift may reach the site in two ways: Firstly, spray drift can directly be deposited in the inlet, the collector shaft, or the stream. This includes overspraying of the site. Secondly, it can reach the site indirectly. In this case, spray drift is deposited on a non-target area (i.e. a road or farm track), and is washed off to the site during the next rain event. We defined spray drift to be possible if the application occurred within less than 100 m from the site (direct spray drift), or from a road or farm track draining to the site (indirect spray drift). Concentrations in this category may originate from spray drift or a process mentioned in the previous categories.

D) *Surface runoff / (tile drainage) / spray drift / other.* This category was assigned if the pesticide was applied before the event and surface runoff to the sampling site was possible. This was defined to be the case if the application occurred within the surface runoff contributing area of the site (determination – see Sect. 4.2.4.1). Concentrations in this category may originate from surface runoff or processes mentioned in the previous categories. For the sites CS and ST, concentrations in this category may also originate from tile drainages.

Although it would have been desirable to further disaggregate the above-mentioned categories (e.g. surface runoff is a possible pathway, but spray drift is not), the spatio-temporal patterns in the study catchment did not allow for such a disaggregation. For example, there were no applications with the potential for surface runoff to a sampling location, but without spray drift potential.

As mentioned previously, for 4% of the agricultural area no application data could be obtained. Since all concerned fields were situated far away from the sampling sites, the influence of the missing application data on our results can be neglected.

4.2.4.4 Discharge transported through inlets

As mentioned in Sect. 4.2.2.2, discharge in the inlets was calculated using water level measurements and a weir with a calibrated rating curve. The rating curve could only be calibrated for water levels corresponding to discharges of up to approximately 0.5 L/s. For higher water levels, we therefore calculated a minimum (Q_{min}), a moderate (Q_{mod}), and a high (Q_{high}) discharge estimate (details – Sect. S4.1.3.2). For the discharge measured in the stream Q_{stream}, no information on uncertainty was provided by the cantonal authorities. Expecting that the relative uncertainty of the discharge through inlets is much larger than the uncertainty in stream discharge, we neglected the latter.

To compare the discharge in the inlets and the stream, we calculated the ratio ($r_{Q,min}$, $r_{Q.mod}$, $r_{Q,high}$) between the discharge estimate sums of all four inlets (Q_{min}, Q_{mod}, Q_{high}) and the discharge in the stream (Q_{stream}) (eq. S4.7). Additionally, we calculated the ratio ($r_{Q,fast,min}$, $r_{Q.fast,mod}$, $r_{Q,fast,high}$) between the discharge estimate sums of all four inlets (Q_{min}, Q_{mod}, Q_{high}) and the fast discharge estimates in the stream ($Q_{stream,fast,high}$, $Q_{stream,fast,mod}$, $Q_{stream,fast,low}$) (eq. S4.8).

The fast discharge in the stream was estimated using a recursive filter technique (Lyne and Hollick, 1979) for discharge separation (function "BaseflowSeparation" of the R package "EcoHydRology", version 0.4.12.1, Fuka et al. (2018)). We used three different filter parameters (0.9, 0.925, and 0.95; see Nathan and Mcmahon (1990)) to come up with a low, moderate, and high estimate of the fast discharge.

Using the discharge measurements in the four inlets, we estimated the total discharge flowing through all inlets in the catchment $Q_{inl,tot}$. For this, we used three simple extrapolation methods. In the first two methods, we assumed that the discharge in an inlet is proportional to the road area (eq. S4.9) or the agricultural area connected to the inlet (eq. S4.10). In the third method, we assumed that the discharge is proportional to the number of inlets (eq. S4.11). These three methods are meant to provide a rough estimate of the total discharge and other parameters influencing the total discharge (such as slope, soil permeability, crop types, spatial distribution of rainfall) were not taken into account.

4.2.4.5 Pesticide loads transported through inlets

To compare pesticide transport in the sampled inlets and the stream, we calculated pesticide loads and their ratio between the inlets and the stream. These calculations were only performed for events with sufficient temporal sampling resolution in the stream, i.e. events 5, 6, and 12, but not events with backup samples from cantonal authorities (see Sect. 4.2.2). These three events correspond to the highest, fourth highest, and sixth highest of the 19 rain events sampled with respect to pesticide concentration sums measured in the inlets. To account for the uncertainty in discharge measurements and for the uncertainty introduced by the analytical limits of quantification (LOQ), we calculated minimum, moderate, and high estimates of the pesticide loads f (eq. 4.1).

$$\mathbf{f_{i,e,s}} = \begin{pmatrix} \mathrm{f_{i,e,s,min}} \\ \mathrm{f_{i,e,s,mod}} \\ \mathrm{f_{i,e,s,high}} \end{pmatrix} = \begin{pmatrix} \mathrm{Q_{i,e,min}} \\ \mathrm{Q_{i,e,mod}} \\ \mathrm{Q_{i,e,high}} \end{pmatrix} \cdot \begin{pmatrix} \mathrm{c_{i,e,s,min}} \\ \mathrm{c_{i,e,s,min}} \\ \mathrm{c_{i,e,s,max}} \end{pmatrix} \tag{4.1}$$

with: $$c_{i,e,s,min} = \begin{cases} c_{i,e,s} \,|c_{i,e,s} \geq LOQ_s \\ 0 \,|c_{i,e,s} < LOQ_s \end{cases}$$

$$c_{i,e,s,max} = \begin{cases} c_{i,e,s} \,|c_{i,e,s} \geq LOQ_s \\ LOQ_s \,|c_{i,e,s} < LOQ_s \end{cases}$$

$f_{i,e,s,min}$, $f_{i,e,s,mod}$, $f_{i,e,s,high}$: Load estimates (ng) of substance s during event e at location i

$Q_{i,e,min}$, $Q_{i,e,mod}$, $Q_{i,e,high}$: Estimates of the total discharge (L)

$c_{i,e,s,min}$, $c_{i,e,s,max}$: Minimal and maximal concentration of substance s (ng L^{-1})

LOQ_s: Limit of quantification of substance s (ng L^{-1})

From these estimates we calculated the ratio between the loads measured in the four inlets and in the stream r_f for each substance and event (eq. 4.2).

$$\mathbf{r_{f,e,s}} = \begin{pmatrix} r_{f,e,s,min} \\ r_{f,e,s,mod} \\ r_{f,e,s,high} \end{pmatrix} = \begin{pmatrix} \frac{\sum_{i=1}^{4} f_{inl,i,e,s,min}}{f_{stream,e,s,high}} \\ \frac{\sum_{i=1}^{4} f_{inl,i,e,s,mod}}{f_{stream,e,s,mod}} \\ \frac{\sum_{i=1}^{4} f_{inl,i,e,s,high}}{f_{stream,e,s,min}} \end{pmatrix} \tag{4.2}$$

$\mathbf{r_{f,e,s}}$: Load ratio estimates between inlets and the stream (-)

In a next step, we calculated the average of the minimal, moderate and high load ratios between the inlets and the stream using two different approaches. In the first approach, we calculated the mean of the load ratios of each single substance and event ($r_{f,\mu,subst}$; eq. 4.3.1). In the second approach, we calculated the ratio between the substance load sums in the four inlets and in the stream ($r_{f,\mu,sum}$; eq. 4.3.2).

$$\mathbf{r_{f,\mu,subst}} = \frac{\sum_{s=1}^{n_s}\sum_{e=1}^{n_e} \mathbf{r_{f,e,s}}}{n_e \cdot n_s} \tag{4.3.1}$$

$$\mathbf{r_{f,\mu,sum}} = \frac{\sum_{s=1}^{n_s}\sum_{e=1}^{n_e}\sum_{i=1}^{4} \mathbf{f_{inl,i,e,s}}}{\sum_{s=1}^{n_s}\sum_{e=1}^{n_e} \mathbf{f_{stream,e,s}}} \tag{4.3.2}$$

n_s: Number of substances s measured (-)

n_e: Number of events e sampled (-)

In a last step, we used the same extrapolation approach as for the discharge (Sect. 4.2.4.4) to come up with a rough estimate of the pesticide load ratio between all inlets in the catchment and the stream.

4.2.4.6 Model of concentrations in inlets

To better understand which factors influence the pesticide concentrations in inlets, we created a linear mixed model with the measured inlet concentrations $\log_{10}(c)$ as a response variable (function “lmer” of the R package “lme4”, version 1.1.27.1, Bates et al. (2015)). As potential explanatory variables, we chose a set of variables commonly considered important for pesticide transport: Time since application t_{appl}, amount of substance applied $\log_{10}(m_{appl})$, Freundlich adsorption coefficient normalized to organic carbon content $\log_{10}(K_{foc})$, octanol-water partition coefficient $\log_{10}(K_{ow})$, substance half-life in water $DT_{50,\,water}$, substance half-life in soil $DT_{50,\,soil}$, moderate estimate of the discharge in the inlet during the event $\log_{10}(Q_{mod})$, type of potential transport processes involved $p_{transport}$ (see Sect. 4.2.4.3), and the inlet sampled i (details – Table S12). Substance properties were obtained from Lewis et al. (2016). The inlet sampled i was defined as a random factor, all other variables as fixed variables. Since the variables

$\log_{10}(K_{foc})$ and $\log_{10}(K_{ow})$ were strongly correlated, $\log_{10}(K_{ow})$ (i.e. the variable with the lower AIC criterion resulting from single variable deletions) was removed. For the analysis, the dataset was reduced to those 20 substances with substance properties available and at least one application in the contributing area of an inlet (details – Table S13).

4.3 Results and discussion

4.3.1 Surface runoff connectivity

The results of the surface runoff connectivity model (Figure 19) show that around 76% of the agricultural area in the catchment has a surface runoff connectivity to the stream. From this area, 25% is directly connected to the stream, and 75% is indirectly connected via inlets. The four sampled inlets drain around 5.7% of the agricultural area connected to inlets in the study catchment and 2.9% of the roads connected to inlets. The collector shaft drains around half of the agricultural and road area in the catchment that is connected to inlets. The remaining agricultural area (24%) is connected to sink areas. Although the water flowing into these sinks is expected to infiltrate, there might still be a connectivity to the stream via subsurface processes, such as tile drainage or ground water flow.

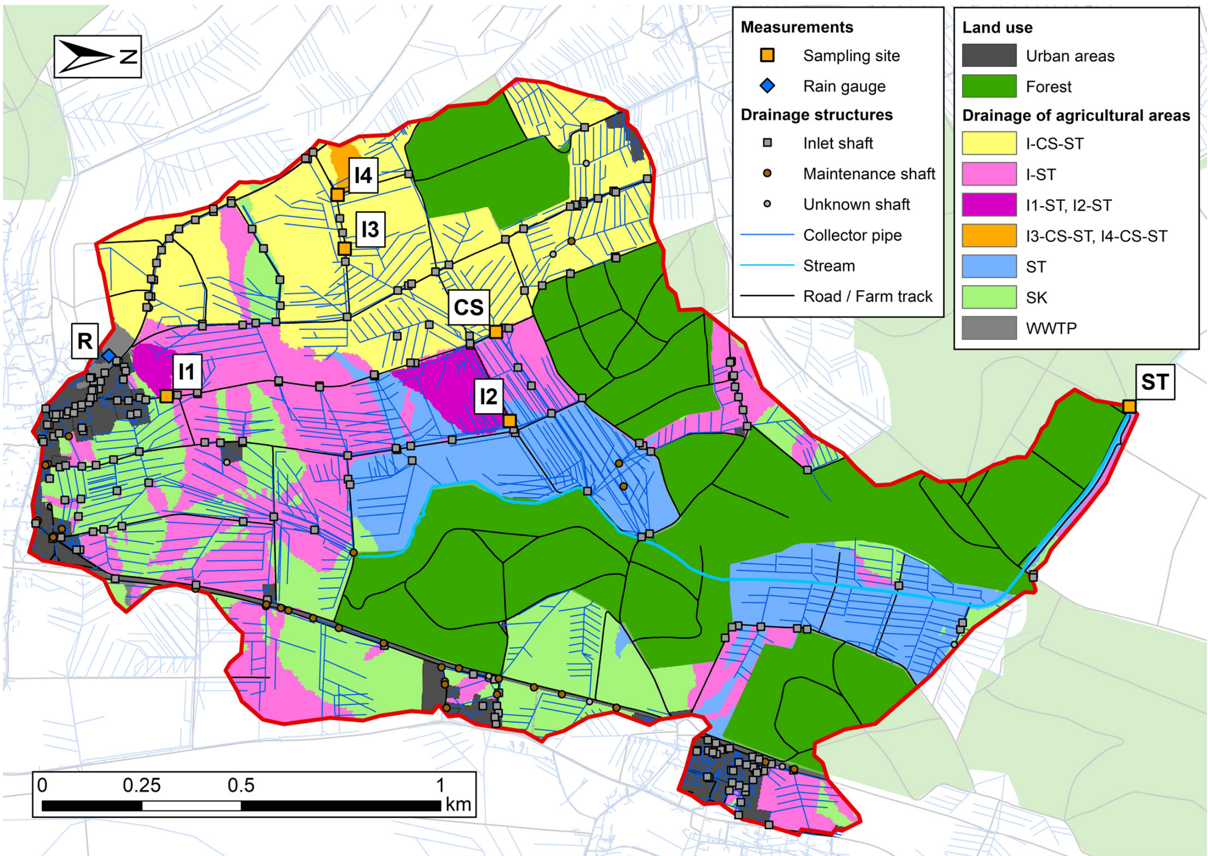


Figure 19: Surface runoff connectivity of the catchment. Yellow squares show the sampling sites (I1-I4: inlets, CS: collector shaft, ST: stream) and the rain gauge (R) is indicated by a blue diamond. Coloured areas show the contributing areas (CAs) of the inlets, sinks, and the stream. I-CS-ST: CAs of inlets draining through the collector shaft into the stream (these inlets were not sampled). I-ST: CAs of inlets draining to the stream without passing the collector shaft (these inlets were not sampled). I1-ST, I2-ST: CAs of inlets 1 and 2, draining to the stream without passing the collector shaft. I3-CS-ST, I4-CS-ST: CAs of inlets 3 and 4, draining through the collector shaft to the stream. (The CA of inlet 3 is small and therefore not visible on the map.) ST: Areas directly drained to the stream. SK: Areas draining to a sink. WWTP: Areas drained to a wastewater treatment plant. Source of background map: Swisstopo (2020b).

These findings are robust when considering the parameter uncertainty of the topographical model. The median area fractions connected to the stream resulting from the Monte Carlo simulation corresponded to 73% of the agricultural areas and the indirect connectivity dominates (83% of the connected agricultural area, or 61% of all agricultural areas). These simulations also allowed us to compare the connectivity of the study catchment to a national connectivity assessment (Schönenberger and Stamm, 2021). The comparison revealed that the study catchment represents conditions with a very high fraction of indirectly connected agricultural area (97 percent quantile of the national distribution). The median of the national distribution (35%) is approximately 1.7 times lower than in the study catchment (61%). Accordingly, we expect that in an average Swiss arable land catchment, surface runoff via inlets and related pesticide transport is lower than in the study catchment, but in a similar order of magnitude.

4.3.2 Hydrological behaviour of inlets

During the study period, 37 rain events were recorded. Their duration was between 1 and 41 hours (median: 9 hours). During 34 rain events, discharge was measured in at least one of the inlets (see Figure S63). The discharge formation in the inlets depended on the total rainfall sum of the respective rain event, but not on the rainfall intensity. The rainfall needed to trigger discharge differed between the inlets. The minimal rainfall sum needed was 1.3 – 1.5 mm for I1, I2, and I4, while I3 was only getting active with 3.6 mm (details Table S14). This can be explained by the grass strip separating I3 from the adjacent road (see Sect. 4.2.2.1). Additionally, due to the seepage through the shaft bottom of I3 during dry periods, surface runoff entering the inlet first had to fill the shaft, before being transported through the outlet pipe. Similarly, the measured discharge differed strongly between the four inlets, being much higher in I1 and I2 than in I3 and I4 (details – Figure S62).

For each rain event, the ratio between the discharge sum of all four inlets and the fast discharge fraction in the stream ($r_{Q,fast}$) is shown in Figure 20. For small events (rainfall < 4 mm), the four inlets are only responsible for less than 0.4% of the fast discharge in the stream. For larger events (rainfall > 10 mm), the contribution is higher with on average 0.83% (0.64 to 1.1%; see Table S15). Event 1 is a clear outlier with $r_{Q,fast}$ equalling around 3.6%. During this event, the ground was covered by melting snow. The snow on the farm tracks was melting faster than on the agricultural areas, explaining the higher discharge transported through the inlets. For small events, the estimation of fast discharge based on discharge separation underlies large uncertainties and should be interpreted with care. A comparison of the discharge sum of all four inlets to the total discharge in the stream (r_Q), revealed similar results with higher contributions of inlets for rain events > 10 mm (details – Figure S64).

The results of the discharge extrapolation from the measured inlets to all inlets in the catchment indicate that for rain events larger than 10 mm, between 3.6% and 10% of the total discharge and between 11% and 43% of the fast discharge in the stream originates from inlets (details – Table S15). These numbers are lower than it would be expected from the connectivity analysis, which estimated that 75% of the areas with surface runoff connectivity are connected to the stream via inlets.. This indicates that the fast discharge in the stream originated to large amounts from other sources than direct and indirect surface runoff from agricultural roads or fields. We hypothesize that preferential flow through tile drainages, surface runoff formed on urban areas, or the fast outflow of pre-event water were major other sources of fast discharge in the stream.

The measurements and extrapolations reported above are only based on measurements in four out of 158 inlets in the catchment. Obviously, the extrapolation to the entire catchment can only provide a very rough estimate of the overall relevance of inlets on the catchment hydrology. In addition, our discharge measurements were restricted to inlets along farm tracks, being the most frequent inlet type in the catchment. Inlets along concrete roads are, however, expected to react much faster (i.e. produce runoff at lower rainfall sums) and to show higher runoff coefficients. In contrast, inlets located directly in fields are expected to react slower and to show lower runoff coefficients. On a national scale, most inlets are located along concrete roads (Schönenberger and Stamm, 2021). We therefore expect that in most other catchments, inlets tend to react faster and to have higher runoff coefficients.

4.3.3 Concentrations and loads

4.3.3.1 Measured concentrations and loads

Inlet water samples were analysed for 19 of 37 rain events, covering 80% of the total discharge transported through the sampled inlets during the study period. In the remaining events, either discharge was too small to trigger sampling (15 events), or no sampling bottles were installed (3 events). Additionally, for six of these events, water samples from the collector shaft and the stream were analysed (details – Table S16). From the 51 substances measured, 43 were found in at least one sample. Between 22 and 33 substances were found in the inlets and the collector shaft, and 42 in the stream (Table 11). The measured concentrations differed strongly between sampling sites. The highest pesticide concentrations were found in I4 for both, mean (291-322 ng/L) and maximal (62000 ng/L, terbuthylazine) concentrations. However, high pesticide concentrations were also found in I1, the collector shaft, and the stream. In contrast, pesticide concentrations in I2 and I3 were much lower. A table with all measured concentrations is provided in SI-B.

The sampling procedure in the inlets (water-level proportional) was different from the one in the collector shaft and the stream (time proportional) which can introduce a bias in the measured concentrations (Bundschuh et al., 2014; Liger et al., 2012; Schleppi et al., 2006). Moreover, the number of events analysed differed between these sites. Therefore, a direct comparison of the concentrations in the inlets to the collector shaft or the stream should be performed with caution. Load calculations – as presented and discussed in Sect. 4.3.3.3 – are more appropriate for a comparison.

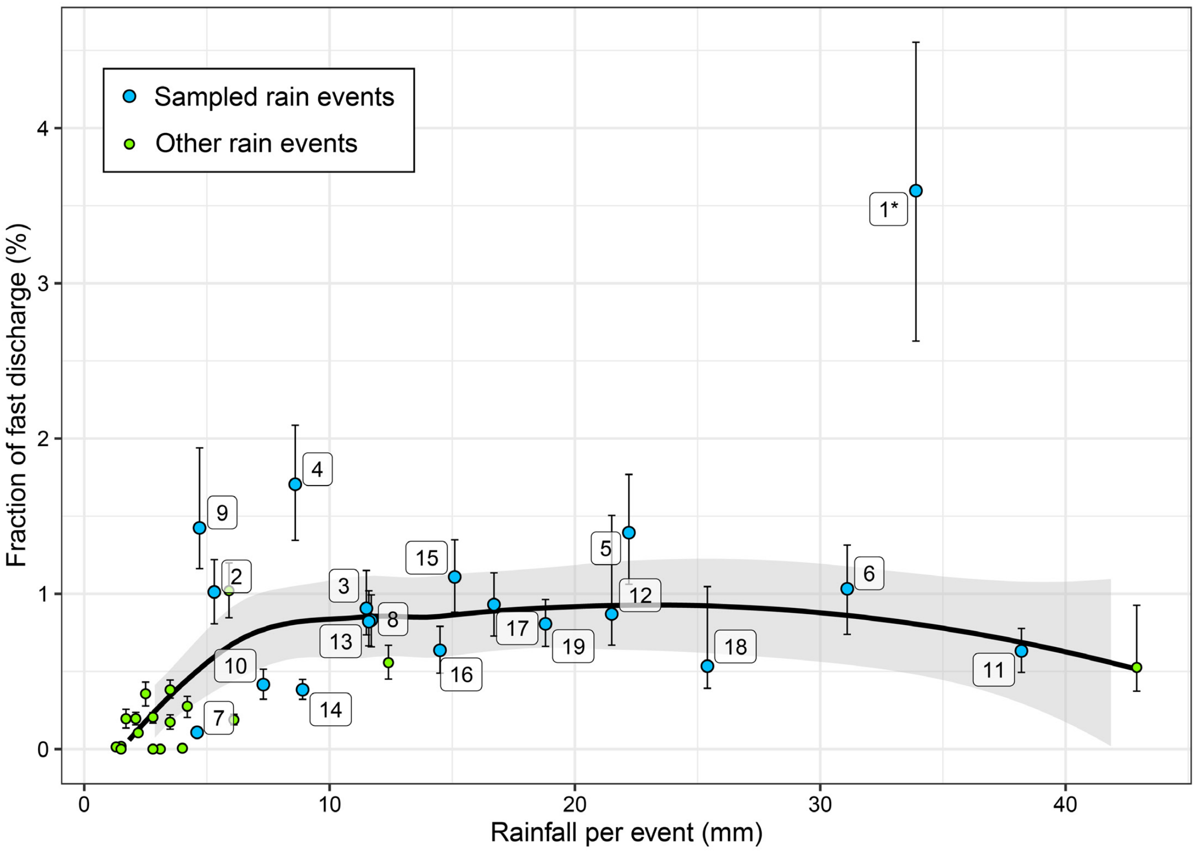


Figure 20: Ratio between the discharge sum in the four inlets and the fast discharge in the stream $r_{Q,fast}$. Points correspond to the moderate estimates ($r_{Q,fast,mod}$), error bars to the minimum and high estimates ($r_{Q,fast,min}$ and $r_{Q,fast,high}$). Sampling event numbers are indicated with white boxes. The numbers represent the events in ascending order of time. The black line represents a smoothed conditional mean of $r_{Q,fast,mod}$, obtained by a locally weighted scatterplot smoothing (LOESS) using the R package ggplot2 (version 3.3.3, function geom_smooth). The grey area represents the corresponding 95% confidence interval. Event 1 was a snowmelt event and was therefore excluded from the analysis.

Table 11: Overview over the pesticide concentrations measured at the different sampling sites. Due to the uncertainty caused by the limit of quantification (LOQ), a range is provided for the mean concentrations. For calculating the lower limit of this range, we replaced the concentrations below the LOQ by zero. For calculating the upper limit, we replaced them by the LOQ. An overview over the measured transformation product concentrations are provided in Table S17. I1-I4: inlets, CS: collector shaft, ST: stream.

Site	I1	I2	I3	I4	CS	ST
Number of substances above LOQ	33	26	22	25	33	42
Mean pesticide concentration (ng L^{-1})	92-124	9-40	11-43	291-322	51-65	190-201
Maximal pesticide concentration (ng L^{-1})	7’900	920	500	62’000	7’900	35’000
Pesticide with highest concentration	Meta-mitron	Meta-mitron	Diflu-fenican	Terbu-thylazine	Terbu-thylazine	Propa-mocarb

In contrast, the concentrations of water-level proportional composite samples in inlets can be compared directly. However, during some events, composite samples were not taken in some inlets, mostly due to lack of sufficient surface runoff (see above). Instead, grab samples from the stagnating water were taken after the event (details – Table S16). Rübel (1999) showed that the pesticide concentrations in surface runoff from vineyard roads are approximately constant within a rain event and that the mixing of different water sources caused the concentration variations observed in the stream. Assuming that this also holds for roads around arable crops, grab sample concentrations can be compared directly to the concentrations of water-level proportional samples, as it is done in the following.

The temporal concentration patterns in the inlets differed strongly between pesticides (Figure 21). Many substances were persistently measured over periods of two months or longer (e.g. metamitron and epoxiconazole at I1, penycuron and metribuzin at I4). This especially holds for substances found in high concentrations. However, other substances were only found in a single sample or two consecutive samples (e.g. propiconazole, cymoxanil, or mecoprop). How these patterns align with pesticide applications and properties is presented in Sect. 4.3.3.2.

Similarly, also the measured loads varied strongly between the inlets and in time. I1 was responsible for the largest fraction of the total load per pesticide transported through the sampled inlets (45%), followed by I4 (30%), I2 (19%), and I3 (6%) (details – Figure S71). Further details on the transported loads are provided in Sect. 4.3.3.3.

4.3.3.2 Factors influencing pesticide concentrations in inlets

Transport processes

We combined the pesticide application data (time, location, substance and amount applied) with the temporal evolution of the concentrations in the inlets. Based on these datasets, we were able to allocate potential transport processes to each measured concentration. This allocation was based on the spatio-temporal relationship between the application and the measured sample, as described in Sect. 4.2.4.3. It allowed gaining insights on the relevance of the different transport processes and other influencing factors on pesticide concentrations in inlets. In Figure 21, the temporal development of the concentrations of the most important compounds is depicted for the 19 sampling events (see Figure S65 and Figure S66 for similar plots for all compounds and the sites CS and ST). Additionally, the respective application timing and potential related transport processes (surface runoff, spray drift, other) are provided. A disaggregated version of this plot with a continuous time axis and including precipitation is provided in the supporting information on the example of epoxiconazole at I1 (Figure S68) and pencycuron at I4 (Figure S69).

These data reveal that applications on fields with surface runoff or spray drift potential to inlets led to strong concentration increases in the corresponding inlets. This was usually observed during the first three events after the application (e.g. bixafen at I1 and I3, terbuthylazine at I4). The highest concentration measured in inlets (terbuthylazine at I4) was related to such an application. Although such a response was not observed in all cases (e.g. metrafenone at I2, cymoxanil at I3), median concentrations in the inlets were clearly related to the potential transport processes (Figure 22). The median concentrations in the inlets decreased from potential surface runoff (category D) over potential spray drift (category C) to other transport processes related to pesticide applications in the catchment (category B), and finally other transport processes not related to a pesticide application in the catchment (category A). This pattern was not only found for pesticides, but also for transformation products. A similar concentration decrease between transport process categories was found in the collector shaft and in the stream.

In summary, high pesticide concentrations in the inlets can be explained in many cases by prior applications on fields with surface runoff or spray drift potential to the corresponding inlet. However, also applications on fields without the potential for these processes to occur led to high concentrations in inlets of up to 7900 ng/L (e.g. metamitron and ethofumesate at I1, propamocarb at I4). The same holds for substances with no application at all reported in the catchment before the respective event (e.g. napropamide and isoproturon at I1, chlortoluron at I1-I4; maximal concentrations up to 1800 ng/L). These results show that also other mechanisms besides surface runoff and spray drift were responsible for high concentrations in inlets. These mechanisms may involve droplet losses, accidental spills, residual wash off from applications in previous years, unreported applications, applications outside the study catchment, or (only in case of I1) farmyard runoff.

The highest concentrations related to applications on fields without surface runoff or spray drift potential were measured in I1 (metamitron and ethofumesate). By rechecking with the farmers, we could exclude unreported applications to be responsible for these concentrations. Additionally, metamitron and ethofumesate have a rather fast degradability ($DT_{50,soil}$: 19 and 22 days; $DT_{50,water}$: 11 and 20 days) and were not applied in the contributing area of the inlet in the year before this study, speaking against wash off of residuals as a source. However, I1 is located close to a village at a farm track often used by farmers for accessing their fields in or outside the study catchment. In contrast, the other inlets are located along farm tracks less often used. This indicates that droplet losses from leaking spraying equipment or accidental spills on the farm track could be responsible for the increased concentrations in I1.

Also in the other inlets, certain substances with rather high degradability ($DT_{50,soil}$ < 25 days) were found in elevated concentrations > 100 ng/L without related applications with surface runoff or spray drift potential (e.g. prosulfocarb at I2, ethofumesate at I4). This again indicates that for some substances droplet losses or accidental spills (but potentially also unreported applications) are responsible for high concentrations in inlets. Contrarily, also substances with low degradability ($DT_{50,soil}$ > 270 days) were measured in elevated concentrations in inlet samples without related applications with surface runoff or spray drift potential (e.g. fluopicolide at all inlets, napropamide at I1). These concentrations likely originated from residual wash off from applications in previous years.

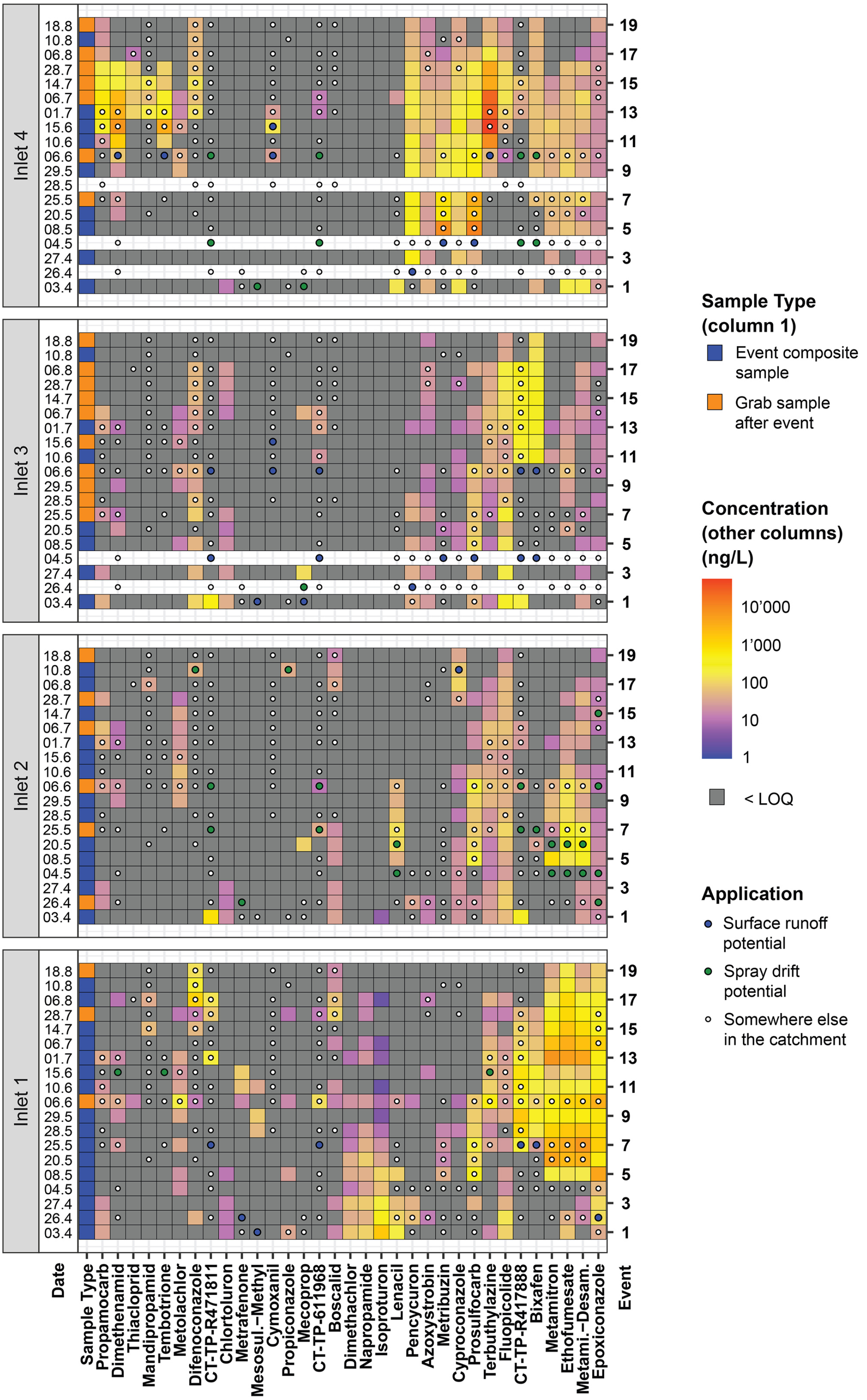


Figure 21: Concentrations c (ng/L) measured in inlets for event 1 (3 April 2019) to 19 (18 August 2019). Only substances found at least twice in concentrations > 25 ng/L are shown. White rows indicate that no sample was taken. In the first column, the sample type is indicated. In the remaining columns, substances are clustered by the concentrations measured. Coloured dots indicate that the particular substance was applied in the period between the respective and the previous event. Dot colours specify the potential transport processes.

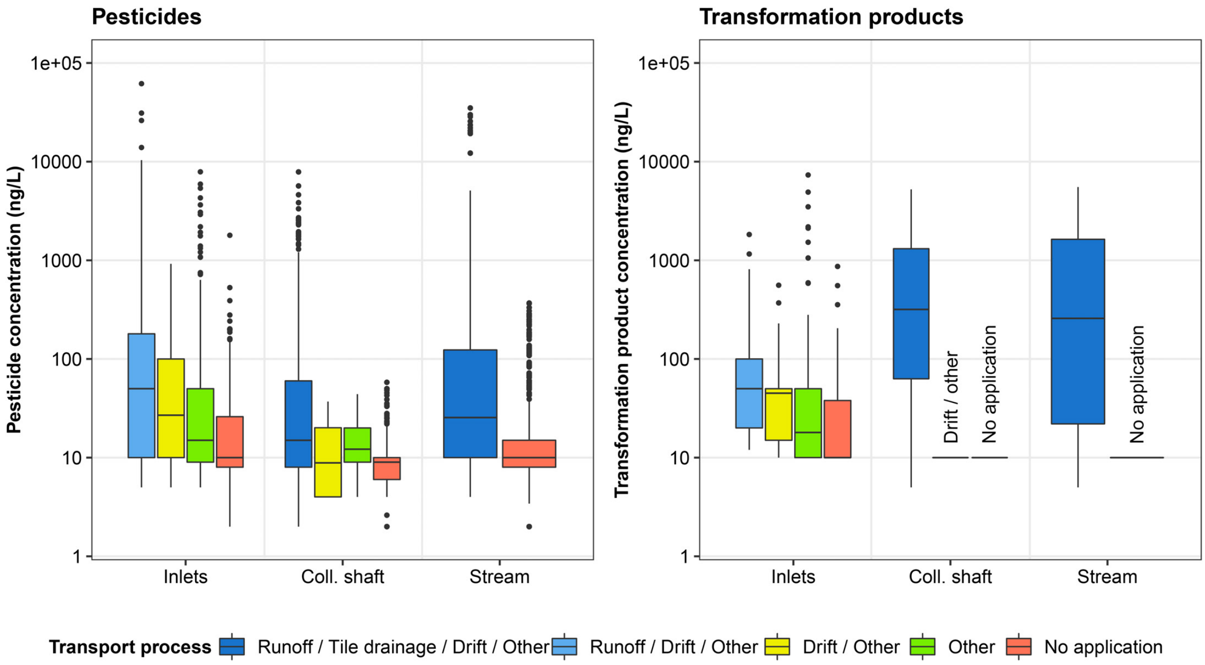


Figure 22: Distribution of pesticide and transformation product concentrations for the sampled inlets, the collector shaft, and the stream. Concentrations are assigned to possible responsible transport processes. For substances below the limit of quantification (LOQ), the LOQ was used for the analysis. A more detailed version of this plot, showing each inlet separately, is provided in Figure S70.

In summary, high pesticide concentrations in inlets are mainly caused by the following transport processes: applications with the potential for surface runoff or spray drift, and potentially droplet losses from leaking spraying equipment or accidental spills on the farm track. This aligns well with studies performed for surface waters, where the same processes have been shown to cause high pesticide concentrations (Holvoet et al., 2007; Reichenberger et al., 2007).

Other influencing factors

The influence of transport processes on the pesticide concentrations in inlets is also shown in the results of the linear mixed model. From all variables tested, the strongest effects on concentrations were observed for the potential transport processes $p_{transport}$.

However, also other factors strongly influenced pesticide concentrations in inlets (details – Table S18). For substances applied on fields with surface runoff or spray drift potential to inlets, high concentrations in the inlets were significantly related to substances with low degradability ($DT_{50,soil}$: $p < 0.001$, $DT_{50,water}$: $p < 0.005$). Such persistent substances are commonly found in streams during dry weather (Halbach et al., 2021; Hermosin et al., 2013; Kreuger, 1998) and can be explained by delayed tile drainage or ground water flow (Gramlich et al., 2018; Reichenberger et al., 2007). However, even though tile drainage and ground water flow cannot enter the inlets, substantial single pesticide concentrations (up to 26000 ng L^{-1}) were found in the grab samples taken in the inlets after the events (Figure 21). This shows that the stagnating water in the inlets (and possibly eroded soil deposited at the inlet bottoms) acted as a pesticide reservoir. Consequently, after an initial rain event with pesticide input, inlets act as pesticide sources and may even lead to pesticide transport to surface waters during rain events with clean surface runoff. This reservoir effect has previously been shown for natural stagnant water bodies (Ulrich et al., 2021), but also to a lesser extent (much lower concentrations) for constructed wetlands (Imfeld et al., 2021; Maillard and Imfeld, 2014). Constructed wetlands are usually reported to overall reduce pesticide transport to surface waters and are therefore often used as a mitigation measure (Vymazal and Březinová, 2015). It was shown that their capability to retain pesticides increases with their density of plant coverage and their hydraulic retention time (Stehle et al., 2011). Inlets have no plant coverage and only a very short hydraulic retention time. Therefore, if we assume that inlets are a special type of constructed wetland, we expect that their efficacy in reducing pesticide transport to surface waters is low and that they act as a pesticide reservoir instead. This aligns well with the results presented here.

Also the Freundlich adsorption coefficient normalized to the organic carbon content $\log_{10}(K_{foc})$, the amount of substance applied $\log_{10}(m_{appl})$, and the time since application t_{appl} were found to significantly influence the concentrations in the inlets (see Table S18). The Freundlich adsorption coefficient and the time since application were correlated negatively to the concentrations in the inlets, while the amount of substance applied was correlated positively. These variables have been previously reported to be important influencing factors for pesticide transport to surface waters (Boithias et al., 2014; Reichenberger et al., 2007). Consequently, our results indicate that pesticide transport to inlets and to surface waters are affected by the same substance properties.

In contrast to the above-mentioned factors, the discharge transported through the inlets per event did not appear as a significant influencing factor in the model. This aligns well with a study by Imfeld et al. (2020) reporting that the event concentrations at the outlet of a small vineyard catchment were related to the timing of pesticide applications, but not to characteristics of the rain events.

4.3.3.3 Relevance of inlets at the catchment scale

Relevance of sampled inlets

In agreement with the large spatio-temporal variability of pesticide concentrations and loads in the sampled inlets, also their contribution to the overall load in the stream largely differed . This is illustrated by Figure 23, showing the load ratios of each pesticide between the sampled inlets and the stream (r_f) for selected events. In some situations, transport through these inlets contributed considerably to the total load of certain pesticides in the stream: In four cases, 10% or more of the load originated from the sampled inlets. In three of these cases, this load was even caused by a single inlet only. However, 40 out of 93 cases, the sampled inlets were of negligible importance for the load in the stream. Overall, the average load ratio per substance between the sampled inlets and the stream ($r_{f,\mu,subst}$) was approximately

1.8% (0.8% to 3.7%) (details – Table S19). In contrast, the ratio between the load sums of all substances in the inlets and in the stream ($r_{f,\mu,sum}$) equalled approximately 0.3% (0.2% to 0.5%). The difference between these two ratios can be explained by few single substances contributing to large extents to the total load in the stream. For example, in event 12, propamocarb alone was responsible for 56% of the total load in the stream.

The differences between the maximum and minimum estimates of $r_{f,\mu,subst}$ and $r_{f,\mu,sum}$ to their moderate estimates were mainly caused by the analytical LOQ. This analytical uncertainty is responsible for 75% and 92% of the total difference between maximum and minimum estimates to the moderate estimates. The remaining differences are caused by the discharge measurement uncertainty. For reducing the load uncertainty in further studies, the focus should therefore be rather set on using analytical methods with lower LOQs than on improving the accuracy of discharge measurements.

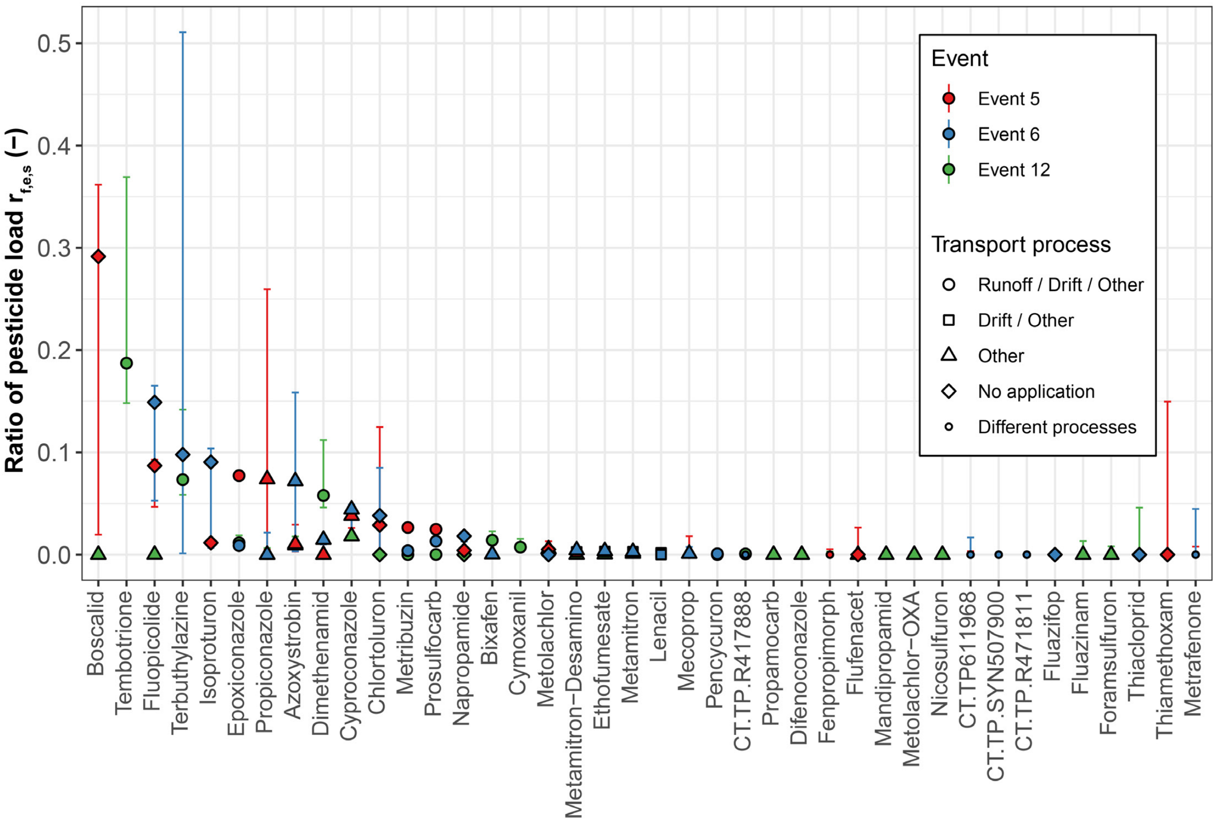


Figure 23: Ratios between the sum of pesticide loads transported through the four sampled inlets and the stream ($r_{f,e,s}$) during selected events (event 5, 6, and 12). Dots represent the moderate estimates ($r_{f,e,s,mod}$), and error bars the minimum ($r_{f,e,s,min}$) and high ($r_{f,e,s,high}$) estimates. Different dot types represent the transport process categories supposed to cause transport to the sampled inlets.

Pesticide load ratios (r_f) were not related to a specific type of potential transport process to the inlets. High pesticide load ratios were found for all transport process types and even for substances without recorded applications (Figure 23). However, high absolute loads (f) were in most cases related to applications in the study catchment (Figure S72). From 46 cases with loads of more than 1 mg in inlets, 20 each were related to a pesticide application with surface runoff potential, and potential for other transport processes only. From the remaining cases, three were related to an application with spray drift potential, and three to either residual wash off from applications in previous years, unreported applications, or applications outside the catchment.

The load ratios reported above were only determined for three rainfall events with rather high pesticide concentration sums measured in the inlets compared to the other events of the study period (see Sect. 4.2.4.5). Likely, the load ratios are therefore smaller for the remaining events.

Besides discharge uncertainty and analytical uncertainty (see above), the different types of sampling methods used (time-proportional in the stream, water-level proportional in inlets) are an additional source of uncertainty in the load calculations. For both methods, the uncertainty related to the sampling method may be substantial if the temporal variations of discharge and concentrations are large within the period covered by a mixed sample. In the stream, however, the temporal sampling resolution was high (see Figure S67 for an example). Therefore, the variation of discharge and concentrations per mixed sample is rather small. Accordingly, we also expect the stream load uncertainty caused by the sampling method to be rather small. For water-level proportional sampling, the influence of temporal variations of discharge and concentrations on the load uncertainty is generally smaller than for time proportional sampling due to the correlation of water level and discharge. As mentioned previously, Rübel (1999) showed that the variation of concentrations on vineyard roads was small during single rain events and stated that a single sample per event is able to represent the event concentration well. Assuming that this conclusion can be transferred to roads around arable crops, we therefore also expect the water-level proportional sampling method to have a small influence on the uncertainty in load calculations.

Relevance of all inlets in the catchment

Based on the load ratios calculated for the sampled inlets and the contributing area characteristics of all inlets, we extrapolated the loads to the entire catchment. We estimate that during the selected events, on average around 30% to 70% of the load of each substance in the stream $r_{f,\mu,subst}$ originated from an inlet in the catchment (details – Table S19). With regard to the load sum ratio $r_{f,\mu,sum}$, we estimate that inlets were responsible for around 5 to 12%.

As already mentioned for the discharge extrapolation, this estimation is only based on measurements in four out of 158 inlets in the catchment. However, substantial differences were found between the loads transported through the four inlets. We therefore suppose that the selection of sampled inlets strongly influenced the load ratios calculated on the catchment scale. For a more robust estimate, additional measurements in other inlets would be essential. Moreover, additional measurements could help to create a more elaborate extrapolation model and to further improve the catchment scale load estimation.

Despite these uncertainties, our results indicate that – at least during some rain events – surface runoff transported through inlets in our study catchment contributed to substantial amounts to the total pesticide load in the stream. Our results are in line with the only other study reporting load ratios for agrochemicals transported through inlets (Remund et al., 2021). In this study, 88% of sediment and phosphorus losses to surface waters occurred through inlets or maintenance shafts. In other countries, storm drainage of fields and adjacent roads is often established by roadside ditches or the roads themselves. In accordance with our results, high pesticide concentrations have been measured in such roadside ditches (Rübel, 1999). Furthermore, in a small agricultural catchment, Louchart et al. (2001) reported that the fast transport of surface runoff via roadside ditches was responsible for 83% and more of the load of two herbicides lost to the stream. In a different catchment, a similar effect was reported for transport via roads (Lefrancq et al., 2014). These results corroborate that structures establishing a surface runoff connectivity between fields and surface waters generally entail a large risk for the transport of substantial pesticide loads to surface waters.

4.3.4 Implications for other catchments

This study was performed in a single catchment and for four inlets only. In the following, we will elaborate which results are rather case-specific and which results can be extrapolated to other catchments.

We found that pesticide concentrations in single inlets can be very high, and that single inlets can be responsible for a large fraction of the pesticide load found in the stream. Assuming that the same processes are driving pesticide transport in other catchments, we suspect that high pesticide transport through inlets may potentially occur in every catchment in which inlets exist and pesticides are applied. If high pesticide concentrations and loads effectively occur in a given inlet, depends on a complex interaction of local influencing factors allowing the above-mentioned transport processes to happen. If pesticide transport is dominated by surface runoff and spray drift, important factors include the spatial arrangement of sprayed crops, roads, and inlets, the local topographical conditions, rainfall patterns, wind conditions, soil and crop types, soil management, type and amount of pesticide applied, and the type of spraying equipment. If pesticide transport is dominated by accidental spills and droplet losses, important factors are the care of farmers during pesticide application and the condition of the spraying equipment.

In Sect. 4.3.3.3, we estimated the ratio of the pesticide load transported through all inlets in the whole catchment during three rain events. This estimation suggests that a very large ratio (30% to 70%) of the pesticide load measured in the stream was transported through inlets. It remains unclear if this ratio is smaller or larger for other catchments and rain events. In the following, we first discuss arguments supporting smaller ratios, and then arguments supporting larger ratios.

The load ratio reported above was calculated for three rain events with rather high pesticide concentrations in the inlets compared to the other events (see Sect. 4.3.3.1). During the other events, we therefore expect the average load ratio to be smaller. Furthermore, compared to an average agricultural catchment in Switzerland, a high fraction of the agricultural area (1.7 times higher than the median) is connected to the stream via inlets in our study catchment (see Sect. 4.3.1). Both considerations indicate that the load ratios reported here are rather case-specific and might on average be smaller for other catchments and rain events.

Contrarily, two different arguments indicate that the average load ratios transported through inlets could be higher in other catchments than the values reported here. First, as mentioned in Sect. 4.3.2, our measurements were performed at inlets located along farm tracks. However, on the national level, most inlets are located along concrete roads. On concrete roads, surface runoff is formed already for very small rainfall events. Therefore, we suppose that on concrete roads the time between pesticide applications and the next rain event causing surface runoff formation is smaller. This could lead to reduced degradation and to increased wash-off of spray drift deposited on roads compared to farm tracks. Second, as mentioned in Sect. 4.2.1, the farmers in the catchment were participating in a program aiming on the reduction of pesticide pollution in the receiving stream. They were aware that transport through inlets might lead to pollution of the stream and that pesticide concentrations are measured in inlets. Thus, especially around the sampled inlets, they were most probably more careful with pesticide handling than farmers in other catchments, leading to lower pesticide transport through the sampled inlets.

4.3.5 Role of application data for process understanding

In many studies conducted on pesticide transport on the catchment scale, application data are not available at all, only in aggregated form, or with other limitations (Hunt et al., 2006; Zhan and Zhang, 2014). Full data sets are often difficult to obtain since the consent and cooperation of all farmers in the catchment is needed, and privacy protection has to be ensured. For this study, we received an almost full dataset of pesticide applications in the study catchment. Even though we were only allowed to report the application data in a aggregated form to ensure privacy protection, our study highlights that linking measured pesticide concentrations to transport processes is only possible given the simultaneous availability of sufficiently resolved application data (plot resolution, daily scale) and sampling data (single inlets, event scale). Without such data, we would have been unable to identify the importance of the different pesticide transport mechanisms in the study catchment or the relevance of compound properties. Moreover, we likely would have confused mechanisms of category B (other processes) with category C or D (surface runoff or spray drift). Consequently, studies aiming to improve the understanding of pesticide transport processes in agricultural catchments should put effort into simultaneously collecting application and sampling data of sufficient spatio-temporal resolution.

Nevertheless, our study also shows that even with available high-resolution application and sampling data, some of the pesticide transport processes had to be suspected (e.g. droplet losses or accidental spills on farm tracks). This illustrates that the pesticide transport processes in agricultural catchments are still poorly understood.

4.4 Conclusions

In this study, discharge and pesticide concentrations were measured for the first time in inlets agricultural storm drainage systems. These inlets were shown to strongly influence surface runoff and related pesticide transport in the studied catchment: The concentrations of single pesticides in inlets amounted up to 62 µg/L and during some rain events, single inlets were responsible for more than 10% of the load of a certain pesticide in the stream. In a rough extrapolation, we estimated that inlets were responsible for 3.6% to 10% of the total discharge in the stream, and for 11% to 43% of the fast discharge fraction. For a subset of three selected large rain events 30% to 70% of the average load per pesticide in the stream originated from inlets. These pesticide load ratios are however rather case-specific and it is difficult to say if the load ratios in other catchments are larger or smaller. To determine which ratio of pesticide pollution in streams originates from inlets, further studies in other catchments are therefore inevitable. Nevertheless, a comparison to other studies suggests that structures increasing the surface runoff connectivity from fields and adjacent roads to surface waters (e.g. inlets, roadside ditches, roads) generally entail a high risk for pesticide loads to surface waters.

This study also provided insights into the processes leading to increased concentrations in inlets. High concentrations were often related to recent pesticide applications on fields with surface runoff or spray drift potential to the sampled inlets. However, increased concentrations in inlets were also found in other cases. Our results indicate that droplet losses or accidental spills on farm tracks may have caused those increased pesticide concentrations. The amount of substance applied, the time since application, and substance properties (DT_{50soil}, $DT_{50water}$, K_{foc}) were identified as other variables with a significant influence on the pesticide concentrations in inlets.

In summary, we conclude from this study that pesticide transport through storm drainage inlets can be a relevant pathway for pesticide pollution of surface waters. This transport pathway should therefore receive more attention in future research, but also in pesticide registration and legislation, and during the application of pesticides.

Chapter 5. Key findings and outlook

Previous research suggested that hydraulic shortcuts (i.e. roads, farm tracks, storm drainage inlet shafts, maintenance shafts, channel drains, ditches) might be an important pathway for pesticide pollution of Swiss surface waters that has been largely overlooked by research, authorities, and farmers. This thesis therefore investigated how relevant such hydraulic shortcuts are for pesticide transport to Swiss surface waters. The following research questions were unknown and were addressed within this thesis: 1) How often do hydraulic shortcuts occur in Switzerland? 2) What is the relevance of indirect surface runoff for pesticide transport? 3) What is the relevance of indirect spray drift for pesticide transport? 4) What pesticide concentrations and loads are transported through hydraulic shortcuts? In the following, the answers to these questions are shortly summarized (Sect. 5.1) and an overarching conclusion is provided (Sect. 5.2). Afterwards, implications for practice (mitigation options, Sect. 5.3) and for research (future research, Sect. 5.4) are discussed.

5.1 Summary

1) How often do hydraulic shortcuts occur in Switzerland?

Before this thesis, it was well known that many roads and farm tracks are located in Swiss agricultural areas. However, it was unknown how many other hydraulic shortcuts (i.e. inlet shafts, maintenance shafts, channel drains, ditches) occur at the national scale. These other shortcuts types were therefore mapped in twenty catchments representing arable land in Switzerland (chapter 2). The results of this mapping campaign showed that:

- Inlet shafts and maintenance shafts are frequently found on arable land areas, while channel drains and ditches occur less often. Inlet shafts were identified as the main shortcut type.
- The majority of inlet shafts is located along roads and farm tracks (90%) and few inlet shafts are located directly in the field (3%).
- With very few exceptions, all of these inlet shafts create connectivity to surface waters via the underground pipe system, either directly (87%) or via wastewater treatment plants and combined sewer overflows (12%).
- For vineyards, the results of chapter 3 suggest that the occurrence of shortcuts is even higher than for arable land.

2) What is the relevance of indirect surface runoff for pesticide transport?

To assess the relevance of indirect surface runoff (i.e. surface runoff transported via hydraulic shortcuts) for pesticide transport, surface runoff connectivity was modelled for twenty catchments representing arable land in Switzerland (chapter 2). The model results show that:

- For around half (47% to 60%) of the arable land areas from which surface runoff can reach surface waters, the connectivity to surface waters is created by hydraulic shortcuts.

- From the surface runoff formed on arable land reaching surface waters and from the related pesticide load, approximately the same fractions are transported via hydraulic shortcuts and via direct runoff.
- For other crop types, the relevance of indirect surface runoff is expected to be different. For example, a higher relevance in vineyards is expected due to their different spatial structure (e.g. higher road drainage densities and steeper slopes) and due to higher pesticide use.

3) What is the relevance of indirect spray drift for pesticide transport?

To assess the relevance of indirect spray drift (i.e. spray drift is deposited on roads and washed-off during subsequent rain events), spray drift was modelled for a representative set of arable land and vineyard catchments (chapter 3). The results show that:

- The amount of spray drift deposited on roads and farm tracks draining to surface waters is much larger than the spray drift directly deposited in surface waters, assuming that farmers comply with the legally required buffer distances.
- Based on current knowledge on pesticide wash off from hard surfaces, major fractions of the drift deposited on roads and farm tracks could be washed off to surface waters, especially in vineyards and for pesticides with low soil adsorption coefficients. In these cases, indirect spray drift may be a major pathway for pesticide losses to surface waters. However, additional research is needed to better quantify the fate of spray drift deposited on roads and farm tracks.

4) What pesticide concentrations and loads are found in hydraulic shortcuts?

In this thesis, pesticide concentrations in inlet shafts were measured for the first time (chapter 4). The results show that:

- High pesticide concentrations and loads can be transported through agricultural storm drainage inlets. Storm drainage inlets strongly influence the transport of surface runoff to the stream and the related pesticide transport.
- High concentrations in inlets were likely related to the following processes: indirect surface runoff, indirect spray drift, or improper handling of pesticides (droplet losses to farm tracks from leaking spraying equipment or due to accidental spills).

5.2 Conclusions

The initial hypothesis of this thesis was that hydraulic shortcuts are an important pathway for pesticide losses to surface waters that has been overlooked in the past. All the above-mentioned answers to this thesis' research questions support this hypothesis. Consequently, transport via hydraulic shortcuts is an important pathway for the pesticide pollution of Swiss surface waters.

In this thesis, three processes were shown to have the potential for causing large pesticide losses via hydraulic shortcuts:

- *Indirect surface runoff*: Surface runoff is formed on crop areas, flows to a shortcut structure, and is then directed to surface waters.
- *Indirect spray drift*: Spray drift is deposited on roads, farm tracks, or other hard surfaces and is washed-off via hydraulic shortcuts to surface waters during the next rain event.
- *Improper handling of pesticides*: Pesticides are lost to roads, farm tracks, or other hard surfaces due to improper pesticide handling (e.g. leaking spraying equipment or accidental spills), and are washed-off via hydraulic shortcuts to surface waters during the next rain event.

In Swiss pesticide legislation and authorization, the effect of hydraulic shortcuts on pesticide transport is at present not considered. Consequently, current regulations and mitigation measures fall short in addressing the full problem of pesticide losses to surface waters. Pesticide transport via shortcuts should therefore be considered in the pesticide registration process and when designing regulations and mitigation measures. Moreover, the awareness of farmers on this transport process should be built and further research should focus on closing remaining knowledge gaps on hydraulic shortcuts. To support these activities, a list of potential mitigation measures is given in Sect. 5.3, and research gaps for future research are identified in Sect. 5.4.

In other countries, no systematic studies on the occurrence of shortcuts are available, but shortcuts were also shown to be important for connecting fields with surface waters in some regions. For example, this was reported in the region of Alsace (France) (Lefrancq et al., 2013), Lower Saxony (Germany) (Bug and Mosimann, 2011), Baden-Württemberg (Germany) (Gassmann et al., 2012), Rhineland-Palatinate (Germany) (Rübel, 1999), and the regions of upper and lower Austria (Hösl et al., 2012). Contrarily to Switzerland, in these regions, mainly roads and ditches acted as a shortcut while pipes were only reported in the study of Gassmann et al. (2012). The occurrence of shortcuts in these regions shows that the results presented in this thesis could also be of relevance for other European countries and that hydraulic shortcuts should receive more attention from authorities and researchers in these countries.

5.3 Potential mitigation measures

The research presented in this thesis shows that hydraulic shortcuts are an important pathway for pesticide transport to surface waters, but have been largely overlooked in the past. Consequently, to this point, no measures to reduce pesticide transport through hydraulic shortcuts have been proposed or implemented (except from few pilot projects). To fill this gap, a list of potential mitigation measures for the reduction of pesticide losses via hydraulic shortcuts is presented in the following (Table 12). This list was adapted to hydraulic shortcuts from a list of general mitigation measures in a review article by Reichenberger et al. (2007). For this, from the general mitigation measures, the ones were selected that seemed most promising with respect to their potential for a reduction of pesticide transport via hydraulic shortcuts at the catchment scale and with respect to practicability (i.e. implementation and maintenance costs, ease of implementation, influence on farming systems). This list was complemented with measures proposed in chapters 2 to 4 or by cantonal authorities (Kanton Aargau, 2020). For all measures, their expected reduction potential, their practicability, and their risk for increasing pesticide transport via other pathways was classified based on current knowledge. Additionally, it was classified, which type of transport via hydraulic shortcuts could be mitigated with these measures. This can either be indirect surface runoff (IR), indirect spray drift (ID), or indirect losses related to improper handling (IH) (e.g. droplet losses to the road due to leaking spraying equipment, or accidental spills to the road).

The list is meant to be a starting point for discussions and further research on mitigation measures against pesticide transport via hydraulic shortcuts and is not meant to be a conclusive review of all possible mitigation options. In the following, each of the proposed mitigation options is shortly explained.

Changing pesticide input into the system

Application rate reduction. By a reduction of the application rate, pesticide losses via IR and ID can be reduced approximately by the amount by which the rate is reduced (Reichenberger et al., 2007). This is one of the easiest measures to implement. As an additional advantage, the purchase costs of pesticides are reduced with this measure. However, the control of pests, weeds, or diseases might be reduced.

Product substitution. A substitution of applied pesticides by other pesticides with different properties (e.g. lower toxicity, higher degradability, lower mobility) can lead to a high reduction of the risk imposed to surface waters by classical transport pathways (Reichenberger et al., 2007). This measure is also expected to have the same effect for the transport pathways IR, ID, and IH. However, depending on the selected substitute, this measure could also have the opposite effect and the substitute substance should therefore be chosen carefully.

Increasing time available for degradation

Shifting application to earlier or later date. The duration between pesticide application and rain events was shown to strongly influence the amount of pesticides lost to surface waters (Boithias et al., 2014). This thesis showed that this is also the case for transport via shortcuts. Therefore, if an application is planned shortly before a rain event, a shift of applications to earlier or later dates may strongly reduce transport via IR, but potentially also via ID and IH. However, shifting of planned applications bears the risk of insufficient pest, weed, or disease control and should be considered carefully.

Reducing runoff from the field to shortcuts

Conservation tillage. Conservation tillage was proposed as an alternative to plow tillage with the aim of mitigating soil erosion (and related pesticide losses). This method could therefore also be used for mitigating pesticide losses via IR. However, conservation tillage produced inconsistent results with respect to the reduction of runoff-related pesticide losses (Elias et al., 2018) and it is unclear if conservation tillage would be effective in reducing pesticide transport via IR.

Buffer strips between fields and drained roads/other shortcuts. Grassed buffer strips at the edge of fields have been shown to efficiently reduce surface runoff and erosion losses, mainly due to infiltration and sedimentation in the buffer strip (Reichenberger et al., 2007). An installation of such buffer strips at the edge of fields next to drained roads or around other shortcuts would therefore highly reduce IR. At the same time, grassed buffer strips would also reduce ID in the same way as no-spray buffers (see below). Grassed buffer strips are cheap and easy to implement, and their main disadvantage is the loss of available crop area.

Reducing spray drift

No-spray buffers between fields and drained roads/other shortcuts. No-spray buffers have been shown to effectively reduce drift to non-target areas (Brown et al., 2004; Ganzelmeier, 1995). Depending on their width, they may therefore also effectively reduce drift to drained roads and farm tracks, or other drained hard surfaces, and consequently reduce pesticide transport via ID. No-spray buffers are cheap and easy to implement. As a potential disadvantage, no-spray buffers might lead to an increase in weed, pest, or disease pressure due to the untreated fraction of crops.

Drift-reducing nozzles. Drift reducing nozzles have been shown to reduce spray drift to non-target areas (FOCUS, 2007) and therefore also reduce pesticide transport via ID. The amount of drift reduction depends on the type of nozzle used. Changing from standard nozzles to drift reducing nozzles is cheap and can be implemented easily. Due to the larger spray droplets, this method may however bear a risk of insufficient droplet distribution on foliage of treated crops.

Reducing connectivity to the stream

Removal of shortcuts. A straightforward approach for mitigating pesticide transport via hydraulic shortcuts would be to remove the shortcuts by structural changes in the landscape. However, shortcuts usually have an important function for traffic and access to the field (roads and farm tracks), or for water drainage (inlet shafts, maintenance shafts, channel drains and ditches). A removal of these structures would therefore likely lead to adverse effects. If shortcuts with water drainage as a primary function are removed, this may cause increased surface runoff, erosion, and direct transport to surface waters, or may lead to flooding and water logging. Therefore, before shortcuts are removed, such adverse effects and the possible need for accompanying measures (e.g. adaptions of the farming system to higher soil moisture) should be considered.

Replacing open lids. In the year 2020, the canton of Aargau has created an information leaflet on mitigation measures to reduce surface water pollution by nutrients and pesticides via drainage shafts on agricultural areas (Kanton Aargau, 2020). For maintenance shafts (i.e. shafts not fulfilling a drainage function) with open lids (e.g. grid lids) within crop areas, they suggest to replace the lid by a sealed lid. These replacements are cheap, very easy to implement, and are expected to reduce pesticide transport via ID, but also via IH (e.g. due to overspraying of the shaft). However, their overall potential for the reduction of indirect pesticide transport is rather low, since only a minor fraction of shafts with open lids is located on crop areas (chapter 2).

Designing inlets as infiltration basins. As mentioned in Sect. 2.4, storm drainage inlet shafts could be designed as small infiltration basins by ensuring that – instead of stagnating in the inlet – water can infiltrate into the soil through the shaft bottom. During rain events, surface runoff would then first fill the inlet up to the height of the outlet pipe. This would prevent some of the surface runoff from reaching surface waters via the pipe system. Since inlets only allow the storage of a small water volume, this measure likely would only have a relevant effect for small rain events. Therefore, it might have a high effect on ID and IH, but only a minor one on IR. However, further research is needed to prove the effectiveness of this measure, and to assess the related costs. Moreover, this measure could also have adverse side effects by increasing pesticide transport through the soil or through tile drainages.

Infiltration basins for storm water drainage pipe outflows. This measure is based on similar considerations as the previous measure. Instead of being infiltrated in the inlet shaft, surface runoff is directed to an infiltration basin via the pipe system and is infiltrated there. However, also for this measure, no research is available that proves its effectiveness for reducing pesticide pollution of surface waters, or assesses the related costs (Reichenberger et al., 2007). Similar to the previous measure, also this measure could lead to increased pesticide transport through the soil or through tile drainages.

Avoiding improper handling

Information campaigns. Campaigns building the awareness of farmers for pesticide pollution are a common approach to mitigate pesticide pollution in surface waters. Such an information campaign was also running in the study catchment described in chapter 4 during the study period. Information campaigns can strongly reduce pesticide transport to surface waters (especially for point sources) if awareness can be built in sufficient amounts (Fischer et al., 1996). With respect to IR, ID, and IH, information campaigns are therefore expected to have a low to high effect, depending on how many farmers can be convinced.

Filling and cleaning operations on the field / Regular inspection of sprayers. As shown in chapter 4, pesticide losses related to improper handling (accidental spills or droplet losses due to leaking equipment to roads or farm tracks) may be transported to surface waters via hydraulic shortcuts (IH). These losses could be reduced by two measures. Firstly, filling and cleaning operations could be performed directly on fields, which would strongly reduce accidental spills and droplet losses to roads and farm tracks. Secondly, droplet losses due to leaking equipment could also be reduced by regular inspections of sprayers. The latter measure is very easy to implement and would at the same time reduce increased drift losses due to damaged sprayers (Ganzelmeier and Rautmann, 2000). The former measure is however not feasible for all types of sprayers (Reichenberger et al., 2007). Both measures would at the same time also strongly reduce losses via point sources.

Table 12: Mitigation measures for the reduction of pesticide losses via hydraulic shortcuts. The list was adapted to hydraulic shortcuts from a list of general mitigation measures provided in Reichenberger et al. (2007). Transport type: IR – indirect surface runoff, ID – indirect spray drift, IH – indirect losses related to improper handling. In the column "risk shift", measures with a potential risk for a shift of pesticide transport to another pathway are marked.

Mitigation approach	**Mitigation measure**	**Transport type affected**	**Potential for reduction at catchment scale**	**Practicability** (implementation and maintenance costs, ease of implementation, influence on farming systems)	**Risk shift**
Changing pesticide input into the system	Application rate reduction	IR, ID	≈ Percentage of application rate reduction	Easy to implement, less pesticide costs, possible reduced pest/weed/disease control	
	Product substitution	IR, ID	Zero to high	Different costs, different effectiveness in pest/weed/disease control	x
Increasing time for degradation	Shifting application to earlier or later date	IR, ID, IH	Potentially very high, but variable	Easy to implement, possible risk of insufficient pest/weed/disease control	
Reducing runoff from the field to shortcuts	Conservation tillage	IR	Inconsistent results	Easy to implement, possible risk of fungal diseases	x
	Buffer strips between fields and drained roads/other shortcuts	IR, ID	High	Easy to implement, maintenance necessary, loss of crop area	
Reducing spray drift	No-spray buffers between fields and drained roads/other shortcuts	ID	Low to high, depending on buffer distance	Easy to implement, slight loss in yield, possible reduced pest/weed/disease control	
	Drift-reducing nozzles	ID	Medium to very high	Easy to implement, low costs, potential issues with foliage due to droplet size	
Reducing connectivity to the stream	Removal of shortcuts	IR, ID, IH	Possibly high	High effort, possible risk of increased erosion, risk of flooding/waterlogging	x
	Replacing open lids of maintenance shafts	ID, IH	Rather low, depending on the proportion of maintenance shafts with open lids	Easy to implement, low costs	
	Designing inlets as infiltration basins	IR, ID, IH	Possibly high for ID and IH, low for IR	Possibly high installation costs	x
	Infiltration basins for storm water pipe outflows	IR, ID, IH	Possibly high for ID and IH, low for IR	Possibly high installation costs	x
Avoiding improper handling	Information campaigns	IH, IR, ID	Low to high, depending if farmers can be convinced	Low costs, difficult to reach all farmers, continuous effort necessary	
	Filling and cleaning operations on the field	IH	Low to high, depending on proportion of leaking sprayers	High requirements for spraying equipment	
	Regular inspection of sprayers	IH, ID	Low to high, depending on proportion of leaking sprayers	Easy to implement, reduces both drift and point-source inputs	

5.4 Future research

Although this thesis helped to better understand the relevance of hydraulic shortcuts for pesticide transport to Swiss surface waters, various research gaps remain and should be addressed in future studies. These research gaps include:

- The surface runoff connectivity model developed in this thesis (chapter 2) provides valuable insights into surface runoff and pesticide connectivity in Swiss agricultural catchments. However, this model has not been validated with independent measurements on surface runoff and pesticide transport in the field. This issue is extensively discussed in Sect. 2.4.
- In chapter 3, spray drift losses to drained roads are compared to direct spray drift losses to surface waters. Based on few studies that have assessed the fate of pesticides deposited on hard surfaces (Jiang and Gan, 2016; Jiang et al., 2012; Ramwell, 2005; Ramwell et al., 2002; Thuyet et al., 2012), it was concluded that for certain substances major fractions of the spray drift deposited on drained roads may be washed off to surface waters (see discussion in chapter 3). However, the knowledge on pesticide degradation, sorption, and wash-off is rather limited for such surfaces. Further research should therefore address these aspects for a broader spectrum of substances, and for a wider range of environmental conditions. These environmental conditions may include different durations between pesticide applications and subsequent rainfall events, different intensities of solar radiation, different temperatures, and different types of hard surfaces (e.g. asphalt roads vs. farm tracks).
- Actual field measurements of surface runoff, pesticide concentrations, and pesticides loads in hydraulic shortcuts have only been performed in four storm drainage inlets during the field study described in chapter 4. Therefore, some of the results obtained in this field study are specific to the study catchment and to the inlets analysed. For a representative assessment of pesticide transport via hydraulic shortcuts, additional measurements in other catchments and for a larger number of hydraulic shortcuts are needed. Measurements covering a larger number of inlet shafts could be performed with less effort in catchments where no tile drainage system exists or where the storm drainage system is separated from the tile drainage system. In such catchments, instead of in inlet shafts, measurements could be performed at the outlet of the storm drainage system. This would provide data on many inlet shafts at the same time, but without interferences by tile drainage flow.
- Within this thesis, pesticide transport was only assessed for pesticides dissolved in the water phase. However, it has been shown that also large amounts of eroded soil are lost to surface waters via hydraulic shortcuts (Remund et al., 2021). Especially for strongly sorbing pesticides, larger amounts of pesticides may thus be transported through hydraulic shortcuts bound to soil particles (Reichenberger et al., 2007). Therefore, the relevance of shortcuts for sorbed pesticide transport should receive attention in future research.
- Automated methods for identifying hydraulic shortcuts on larger scales could help to systematically determine risk areas for pesticide transport via shortcuts. This could for example be achieved by a combination of high-resolution aerial images with an automated detection algorithm for hydraulic

shortcuts (Mattheuwsen and Vergauwen, 2020; Moy de Vitry et al., 2018). With increasing quality of remote sensing data (resolution of aerial images), such approaches could be used in the future for generating a national connectivity map explicitly considering the locations of hydraulic shortcuts. Such a map could help authorities to identify risk catchments for indirect pesticide losses and could facilitate the advising of farmers.

- In Sect. 5.3, potential mitigation measures for reducing indirect pesticide losses to surface waters are listed. However, for some of these measures it remains unclear by how much they would reduce pesticide losses to surface waters. Further research should therefore assess the effectiveness of reduction measures, but also the related costs and potential disadvantages.
- The relevance of shortcuts on pesticide losses to surface waters has not been systematically assessed in other countries, even though shortcuts were shown to be important for connecting fields with surface waters in some regions (see Sect. 5.2). Also in other countries, shortcuts may therefore have been overlooked and further research should aim on systematically assessing their occurrence and their relevance for pesticide transport.

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S.2. Supporting Information Chapter 2

Hydraulic shortcuts increase the connectivity of arable land areas to surface waters

Datasets & code available on the Eawag Research Data Institutional Repository.

DOI: 10.25678/0003J3

S2.1 Methods

S2.1.1 Catchment statistics

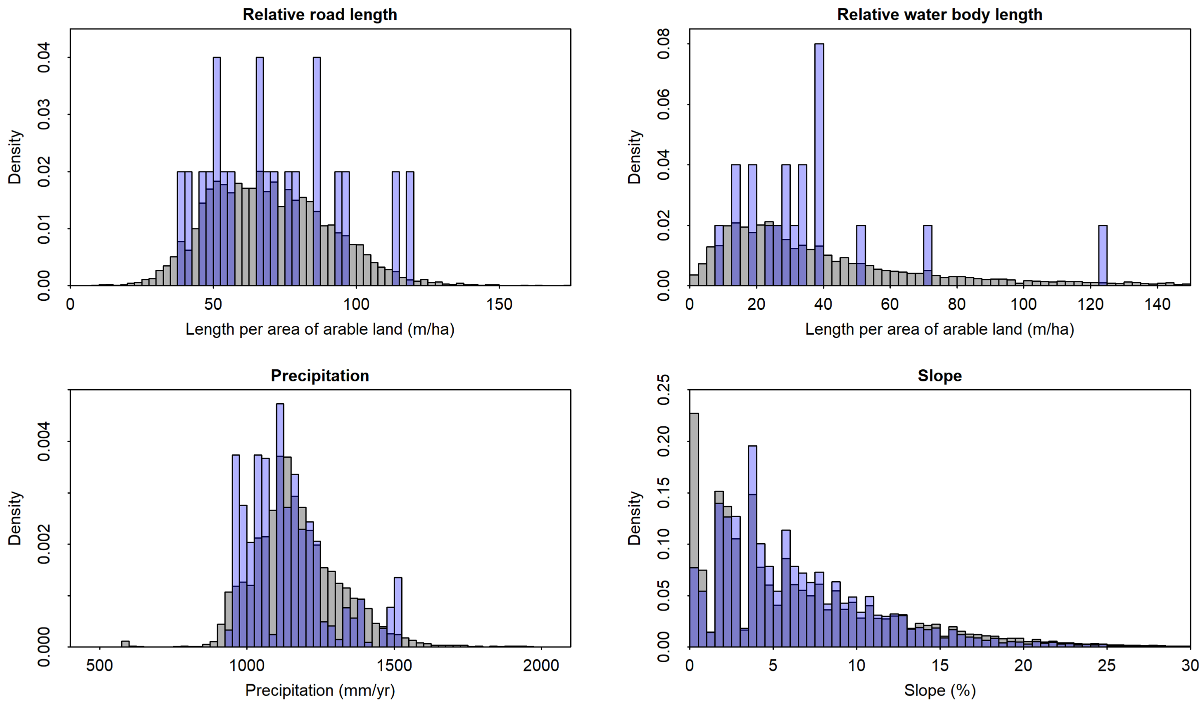


Figure S1: Histogram of catchment statistics for study areas (blue) and all catchments in Switzerland containing arable land (grey). Catchment statistics were calculated only for catchment parts defined as arable land areas by the dataset BFS (2014). Relative road length (road length per arable land area) and relative water body length (water body length per arable land area) were derived from the dataset swissTLM3D (Swisstopo, 2010). Precipitation was derived from Kirchhofer and Sevruk (1992), and slope from Swisstopo (2018).

Table S1: List of catchment statistics calculated for finding explanatory variables for extrapolation to the national scale. Additionally, the datasets used for calculating those statistics are shown.

Catchment statistic	Data source	Dataset used
Fraction of forests	swissTLM3D (Swisstopo, 2010): TLM_BODENBEDECKUNG	OBJEKTART in [12,13]
Fraction of agricultural area	swissTLM3D (Swisstopo, 2010): o TLM_BODENBEDECKUNG, o TLM_STRASSEN, o TLM_SIEDLUNGSNAME, o TLM_NUTZUNGSAREAL	(Total area) - (forests, water bodies, urban areas, traffic areas, and other non-agricultural areas)
Road density (total; paved; unpaved)	swissTLM3D (Swisstopo, 2010): TLM_STRASSEN	BELAGSART in [100,200]; BELAGSART = 100; BELAGSART = 200
Water body density (total; rivers; lakeshores)	swissTLM3D (Swisstopo, 2010): o TLM_FLIESSGEWAESSER o TLM_STEHENDES_GEWAESSER	Both datasets; TLM_FLIESSGEWAESSER only; TLM_STEHENDES_GEWAESSER only
Mean annual precipitation	Kirchhofer and Sevruk (1992)	Mean annual precipitation depths 1951-1980
Mean slope of agricultural areas	swissALTI3D (Swisstopo, 2018)	Slopes as calculated by swisstopo, agricultural areas as defined above
Area fractions (direct; indirect; not connected)	Alder et al. (2015)	Fraction of total directly connected area; fraction of total indirectly connected area; fraction of total not connected area

S2.1.2 Examples of mapped structures

A1 - Storm drainage inlet shafts on or next to roads or farm tracks

Storm drainage inlet shafts on or next to roads or farm tracks were always considered as a potential shortcut in the connectivity model.

Figure S2: Storm drainage inlet shaft with a gridded metal lid on a road in the study area Nürensdorf

Figure S3: Lateral concrete storm drainage inlet shaft next to a road in the study area Molondin

Figure S4: Storm drainage inlet shaft with a gridded metal lid on a road in the study area Oberneunforn

A2 - Strom drainage inlet shafts on fields

Storm drainage inlet shafts on fields are always considered as a potential shortcut in the connectivity model.

Figure S5: Storm drainage inlet shaft with a metal grid lid in a field of the study area Meyrin

Figure S6: Storm drainage inlet shaft with a concrete grid lid in a field of the study area Nürensdorf

B1 – Maintenance shafts on or next to roads

Maintenance shafts on or next to roads are considered a potential shortcut if they are located in an internal sink (only for shortcut definition B).

Figure S7: Maintenance shaft with a metal lid with a pick hole next to a road in the study area Buchs

Figure S8: Maintenance shaft with a concrete lid with a pick hole on a road in the study area Courroux

B2 – Maintenance shafts on fields

Maintenance shafts on fields are considered a potential shortcut if they are located in an internal sink (only for shortcut definition B).

Figure S9: Damaged tile drainage maintenance shaft in a field in the study area Vufflens-la-Ville

Figure S10: Tile drainage maintenance shaft in a field in the study area Molondin

C1 – Channel drains

Figure S11: Channel drain on a road in the study area Clarmont

Figure S12: Channel drain and inlet shaft with a metal grid lid on a road in the study area Lommiswil

C2 – Ditches

Figure S13: Ditch between a field and a road in the study area Meyrin

S2.1.3 List of mapped structures

Table S2: Types of mapped point features

ID	Description	Potential shortcut
1	Inlet shaft	Yes
2	Maintenance shaft	If lying in an internal sink (shortcut definition B)
3	Other shaft	If lying in an internal sink (shortcut definition B)
4	Stormwater tank	If lying in an internal sink (shortcut definition B)
5	Spillway	If lying in an internal sink (shortcut definition B)
6	Pumping station	No
7	House connection	No
8	Other point object	No
9	Unknown shaft	If lying in an internal sink (shortcut definition B)
10	Outfall	No
11	Infiltration structure	If lying in an internal sink (shortcut definition B)
12	Unknown object	No

Table S3: Types of lids

ID	Description
1	Metal grid
2	Concrete lid with pick hole
3	Concrete lid without pick hole
4	Metal lid with pick hole
5	Metal lid without pick hole
6	Other lid type
7	Concrete grid
8	Concrete lid with lateral inlet
9	Metal lid with lateral inlet
0	Unknown lid type

Table S4: Types of line features mapped

ID	Description	Potential shortcut
1	Drainage pipe	No
2	Tile drainage pipe	No
3	Other pipe	No
4	Channel drain	Yes
5	Ditch	Yes
6	Sequence of channel drains & ditches	Yes
7	Stone wall	No
8	Earth wall	No
9	Hedge	No
10	River	No
11	Other line objects	No
12	Unknown line objects	No

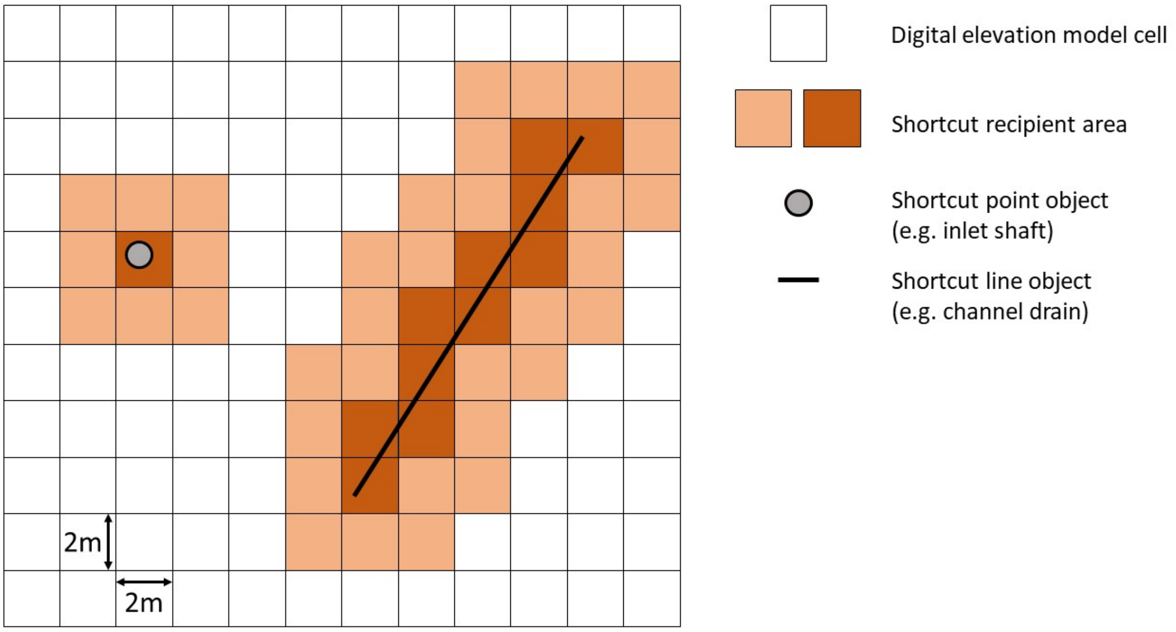


Figure S14: Definition of shortcut recipient areas

S2.1.4 Dates of field mapping and drone flights

Table S5: Dates of field mapping and drone flights for each study area. In some areas a second drone flight had to be performed to ensure sufficient image quality.

ID	Location	Date field mapping	Date drone flights
1	Böttstein	26.10.2017	26.10.2017
2	Ueken	25.10.2017	25.10.2017
3	Rüti b. R.	23.11.2017	23.11.2017
4	Romont	02.11.2017	03.11.2017
5	Meyrin	27.11.2017	Usage of cantonal aerial images only
6	Boncourt	24.11.2017	24.11.2017; 07.06.2018
7	Courroux	17.11.2017	17.11.2017
8	Hochdorf	29.09.2017	27.04.2018
9	Müswangen	21.09.2017	16.08.2018
10	Fleurier	24.05.2018	24.05.2018
11	Lommiswil	16.11.2017	16.11.2017
12	Illighausen	30.08.2017	07.12.2017
13	Oberneunforn	06.09.2017	01.11.2017; 19.04.2018
14	Clarmont	09.11.2017	10.11.2017; 04.12.2017
15	Molondin	02.11.2017	03.11.2017
16	Suchy	10.11.2017	08.11.2017
17	Vufflens	09.11.2017	08.11.2017; 24.08.2018
18	Buchs	23.08.2017	09.08.2017; 17.08.2017
19	Nürensdorf	18.09.2017	24.10.2017
20	Truttikon	20.09.2017	01.11.2017

S2.1.5 Extrapolation to the national scale

In the following, mathematical details on the extrapolation of the local surface runoff connectivity model (LSCM) to the national scale are given. A schematic overview is given in the main part of this publication. Our model is using the area fractions of the national erosion connectivity model (NECM) to extrapolate the LSCM to the national scale, resulting in area fractions of a national surface runoff connectivity model (NSCM).

We defined the area fractions of model *m* and catchment *c* as follows:

$$\mathbf{f_m} = \begin{pmatrix} \overrightarrow{\mathrm{f_{m,dir}}}^{\mathrm{T}} \\ \overrightarrow{\mathrm{f_{m,indir}}}^{\mathrm{T}} \\ \overrightarrow{\mathrm{f_{m,nc}}}^{\mathrm{T}} \end{pmatrix} = \begin{pmatrix} \mathrm{f_{m,dir,1}} & \cdots & \mathrm{f_{m,dir,c}} & \cdots & \mathrm{f_{m,dir,n}} \\ \mathrm{f_{m,indir,1}} & \cdots & \mathrm{f_{m,indir,c}} & \cdots & \mathrm{f_{m,indir,n}} \\ \mathrm{f_{m,nc,1}} & \cdots & \mathrm{f_{m,nc,c}} & \cdots & \mathrm{f_{m,nc,n}} \end{pmatrix}$$

$$= \begin{pmatrix} \frac{\mathrm{A_{m,dir,1}}}{\mathrm{A_{tot,1}}} & \cdots & \frac{\mathrm{A_{m,dir,c}}}{\mathrm{A_{tot,c}}} & \cdots & \frac{\mathrm{A_{m,dir,n}}}{\mathrm{A_{tot,n}}} \\ \frac{\mathrm{A_{m,indir,1}}}{\mathrm{A_{tot,1}}} & \cdots & \frac{\mathrm{A_{m,indir,c}}}{\mathrm{A_{tot,c}}} & \cdots & \frac{\mathrm{A_{m,indir,n}}}{\mathrm{A_{tot,n}}} \\ \frac{\mathrm{A_{m,nc,1}}}{\mathrm{A_{tot,1}}} & \cdots & \frac{\mathrm{A_{m,nc,c}}}{\mathrm{A_{tot,c}}} & \cdots & \frac{\mathrm{A_{m,nc,n}}}{\mathrm{A_{tot,n}}} \end{pmatrix} \tag{S2.1}$$

with:

m:	Model (either LSCM, NECM, or NSCM)
$A_{m,dir,c}$:	Directly connected agricultural area of model m in catchment c (ha)
$A_{m,indir,c}$:	Indirectly connected agricultural area of model m in catchment c (ha)
$A_{m,nc,c}$:	Not connected agricultural area of model m in catchment c (ha)
$A_{tot,c}$:	Total agricultural area in catchment c (ha)
$f_{m,dir,c}$:	Fraction of directly connected agricultural areas of model m in catchment *c* (-)
$f_{m,indir,c}$:	Fraction of indirectly connected agricultural areas of model m in catchment *c* (-)
$f_{m,nc,c}$:	Fraction of not connected agricultural areas of model m in catchment *c* (-)

The area fraction matrices $\mathbf{f_m}$ underlie two boundary conditions (see main part). To ensure that extrapolation model meets these boundary conditions, we used a unit simplex transformation approach.

We performed a unit simplex inverse transformation to the area fraction matrices of the LSCM $\mathbf{f_{LSCM}}$ and the NECM $\mathbf{f_{NECM}}$ (3x20 matrices), resulting in the matrices $\mathbf{z_{LSCM}}$ and $\mathbf{z_{NECM}}$ (2x20 matrices).

$$\mathbf{z} = \begin{pmatrix} \overrightarrow{\mathrm{z_1}}^{\mathrm{T}} \\ \overrightarrow{\mathrm{z_2}}^{\mathrm{T}} \end{pmatrix} = \begin{cases} \mathrm{logit}^{-1}\left(\overrightarrow{\mathrm{f_k}}^{\mathrm{T}} + \log\left(\frac{1}{\mathrm{K-k}}\right)\right) & \mid \mathrm{k} = 1 \\ \left(1 - \sum_{\mathrm{k=1}}^{\mathrm{k-1}} \overrightarrow{\mathrm{z_k}}^{\mathrm{T}}\right) \cdot \mathrm{logit}^{-1}\left(\overrightarrow{\mathrm{f_k}}^{\mathrm{T}} + \log\left(\frac{1}{\mathrm{K-k}}\right)\right) = \left(1 - \overrightarrow{\mathrm{z_1}}^{\mathrm{T}}\right) \cdot \mathrm{logit}^{-1}\left(\overrightarrow{\mathrm{f_k}}^{\mathrm{T}}\right) & \mid \mathrm{k} = 2 \end{cases} \quad \text{with: } \mathrm{K} = 3 \tag{S2.2}$$

In order to model the difference $\Delta\mathbf{z}$ (2x20 matrix) between the transformed LSCM and the transformed NECM ($\Delta\mathbf{z} = \mathbf{z_{LSCM}} - \mathbf{z_{NECM}}$), we tested the same list of nationally available catchment statistics that was already used before. For each of the two dimensions, we selected the variable that correlated best with $\Delta\mathbf{z}$. Those were the fraction of directly connected areas $f_{NECM,dir}$, and the fraction of indirectly connected areas $f_{NECM,indir}$. Using these variables, we performed the following linear regression to describe $\Delta\mathbf{z}$:

$$\Delta\mathbf{z} = \vec{a} + \vec{b} \cdot \begin{pmatrix} \overline{f_{NECM,dir}}^{T} \\ \overline{f_{NECM,indir}}^{T} \end{pmatrix} + \vec{\varepsilon} \tag{S2.3}$$

For each of the catchments of the transformed national erosion connectivity model ($\boldsymbol{z}_{NECM}$, 2xn matrix, n = 11'503), this linear regression was used to calculate the transformed national surface runoff connectivity model ($\mathbf{z}_{NSCM}$, 2xn matrix):

$$\mathbf{z}_{NSCM} = \mathbf{z}_{NECM} + \Delta\mathbf{z} \tag{S2.4}$$

Finally, using a unit simplex transformation, we transformed $\mathbf{z}_{NSCM}$ back, resulting in the area fraction matrix of the national surface runoff connectivity model $\mathbf{f}_{NSCM}$ (3xn matrix).

$$\mathbf{f}_{NSCM} = \begin{cases} \mathbf{f}_{NSCM,k} = \operatorname{logit}(\mathbf{z}_{NSCM,k}) - \log\left(\frac{1}{K-k}\right) & \mid k = 1 \\ \mathbf{f}_{NSCM,k} = \operatorname{logit}\left(\frac{\mathbf{z}_{NSCM,k}}{1-\sum_{k=1}^{k-1}\mathbf{z}_{NSCM,k}}\right) - \log\left(\frac{1}{K-k}\right) & \mid k > 1 \end{cases} \quad \text{with } K = 3 \tag{S2.5}$$

This extrapolation model was run for each of the 100 area fractions matrices resulting from the Monte Carlo analysis that was performed on the local scale.

To address the uncertainty introduced by the selection of our study catchments, we bootstrapped the model 100 times. For each of the bootstrapping iterations 20 of our study catchments were resampled randomly.

S2.2 Results

S2.2.1 Occurrence of hydraulic shortcuts

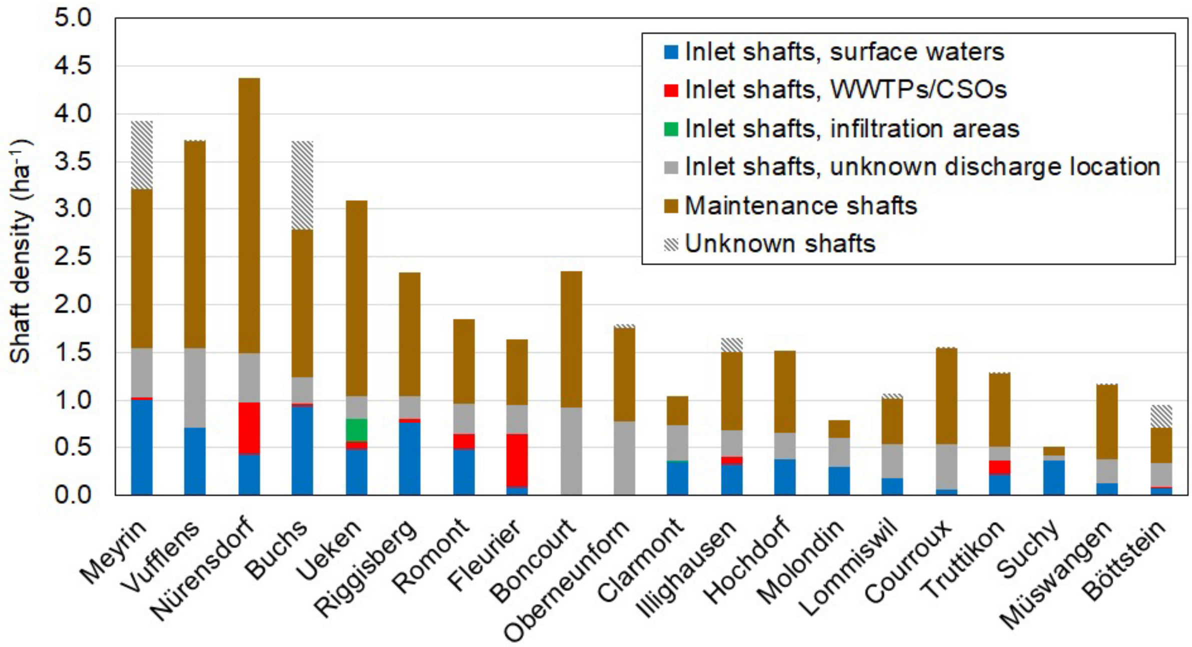


Figure S15: Shaft density (ha^{-1}) on agricultural areas of the study catchments. For inlet shafts, colors show the drainage locations of the shafts. Abbreviations: WWTPs – waste water treatment plants, CSOs – combined sewer overflows.

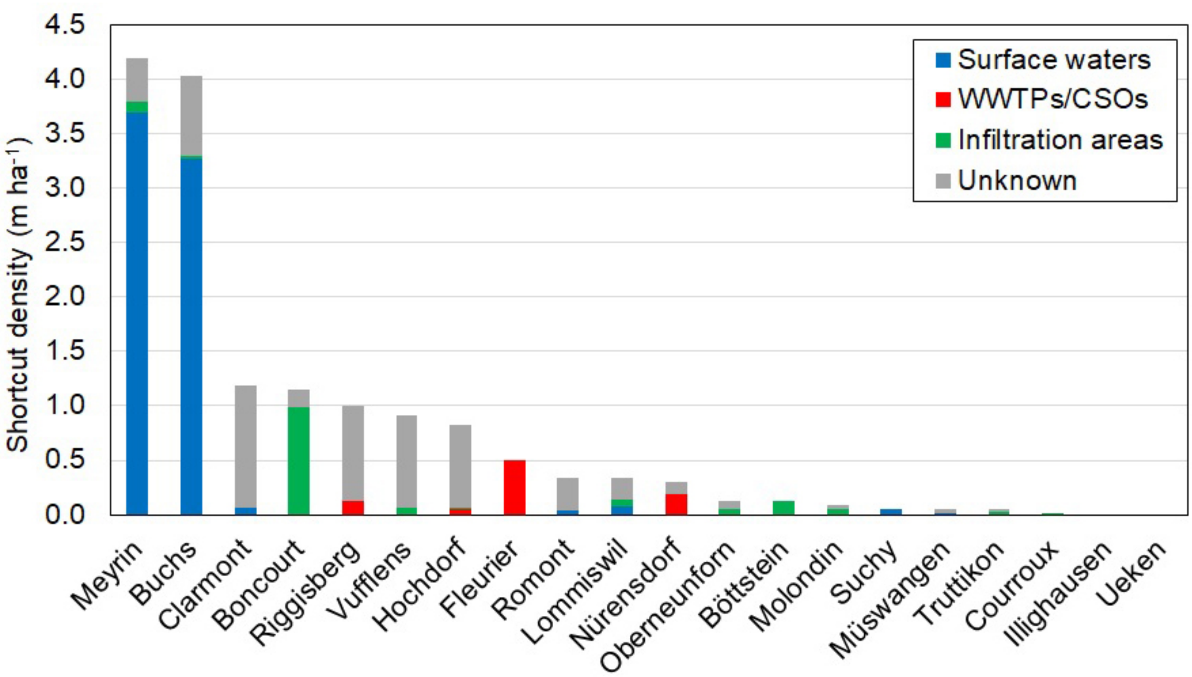


Figure S16: Density of channel drains and ditches (m ha^{-1}) on agricultural areas of the study catchments. Colors show the drainage locations of the channel drains and ditches. Abbreviations: WWTPs – waste water treatment plants, CSOs – combined sewer overflows.

Table S6: Linear regression of different catchment statistics with inlet shaft densities (ha^{-1}) per study area. R^2 equals the coefficient of determination, m is the slope of the linear regression, and p is the p-value.

Catchment statistic	R^2	m	p
Paved road density (m^{-1})	3.3E-01	5.7E+01	8.4E-03**
Unpaved road density (m^{-1})	6.3E-02	-1.5E+01	2.8E-01
Mean annual precipitation (mm yr^{-1})	4.9E-04	-5.1E-05	9.3E-01
Mean slope on agricultural areas (deg)	8.3E-04	-4.7E-03	9.0E-01
Surface water body density (m^{-1})	4.4E-02	-4.3E-05	3.7E-01
Subsurface water body density (m^{-1})	6.2E-02	5.1E+02	2.9E-01

Table S7: Linear regression of different catchment statistics with maintenance shaft densities (ha^{-1}) per study area. R^2 equals the coefficient of determination, m is the slope of the linear regression, and p is the p-value.

Catchment statistic	R^2	m	p
Paved road density (m^{-1})	3.7E-01	1.8E+02	4.6E-03**
Unpaved road density (m^{-1})	3.1E-02	-3.2E+01	4.6E-01
Mean annual precipitation (mm yr^{-1})	4.2E-03	-4.5E-04	7.9E-01
Mean slope on agricultural areas (deg)	1.6E-02	-6.2E-02	6.0E-01
Surface water body density (m^{-1})	3.5E-02	-1.2E-04	4.3E-01
Subsurface water body density (m^{-1})	1.2E-01	2.2E+03	1.3E-01

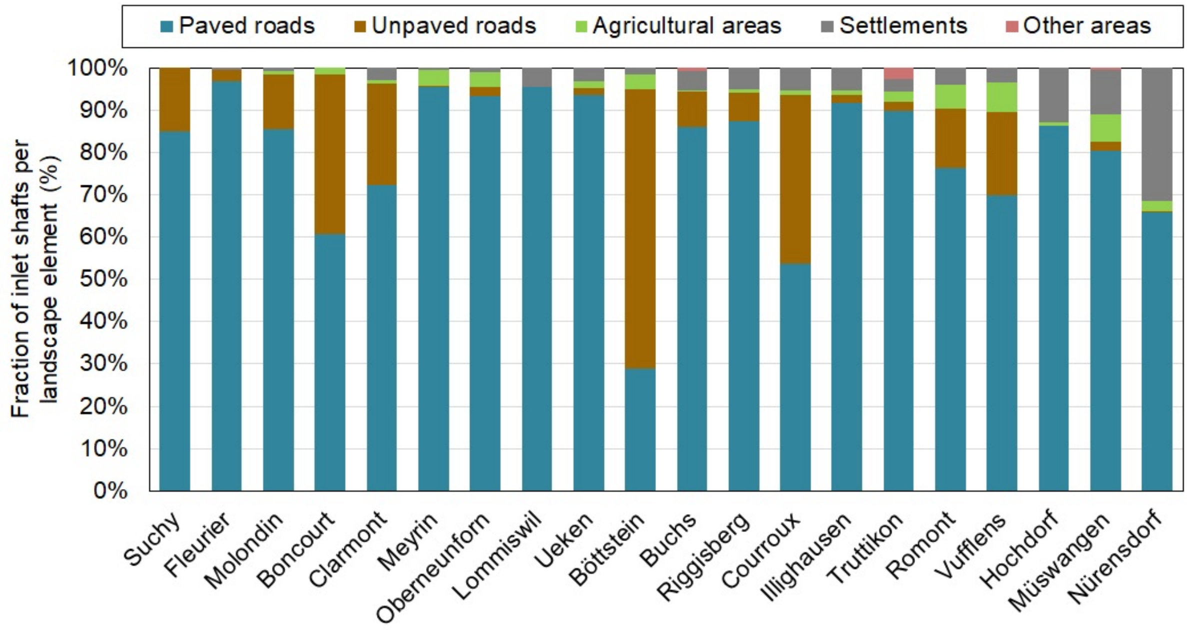


Figure S17: Fraction of inlet shafts per study area belonging to a certain landscape element

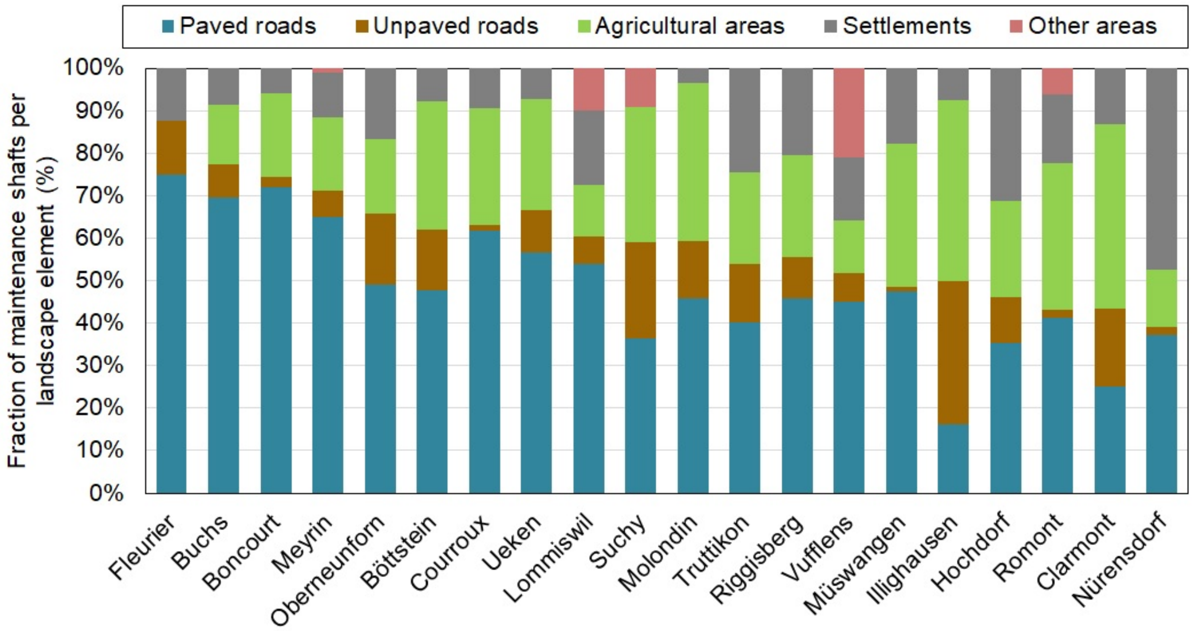
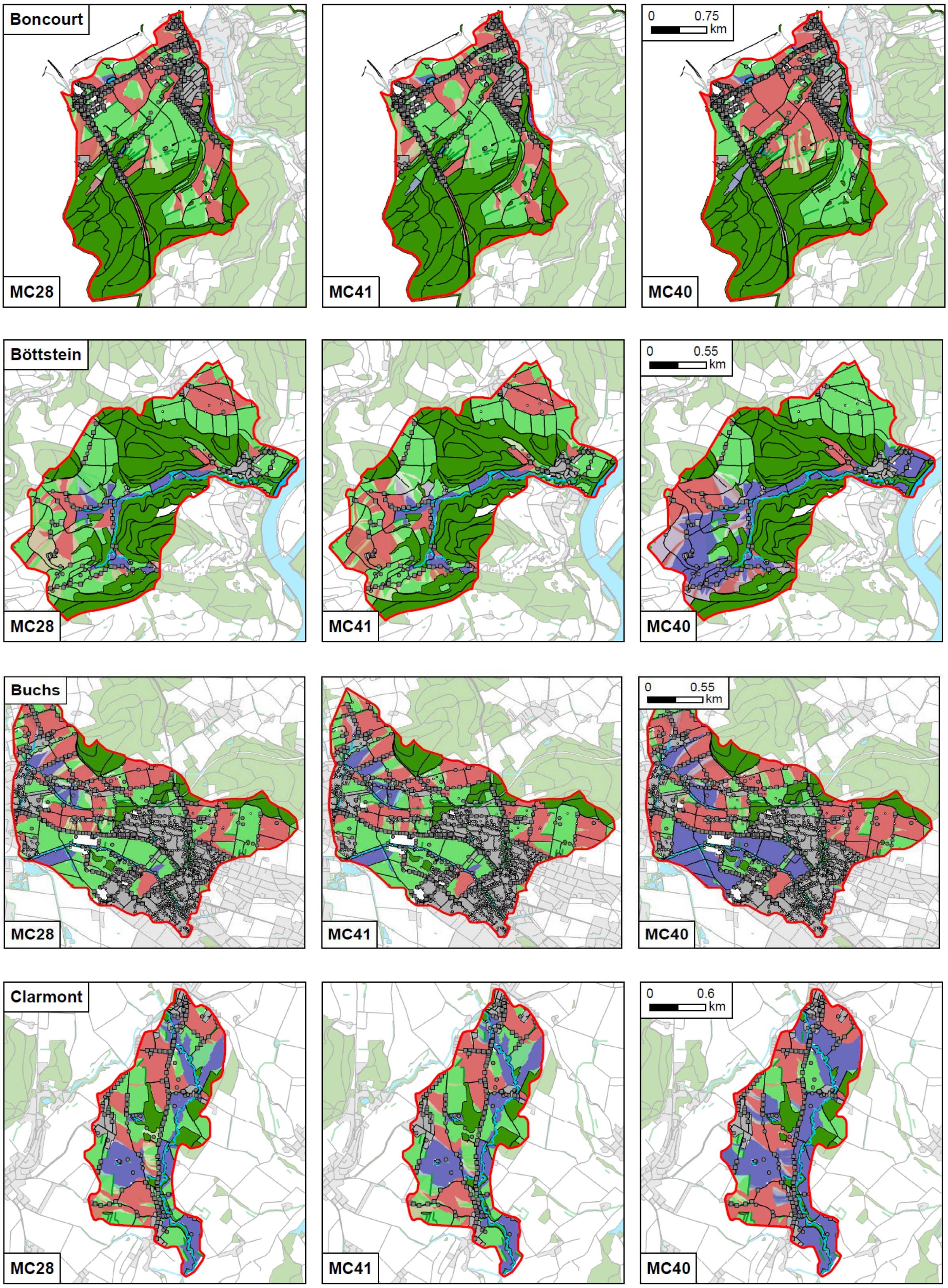


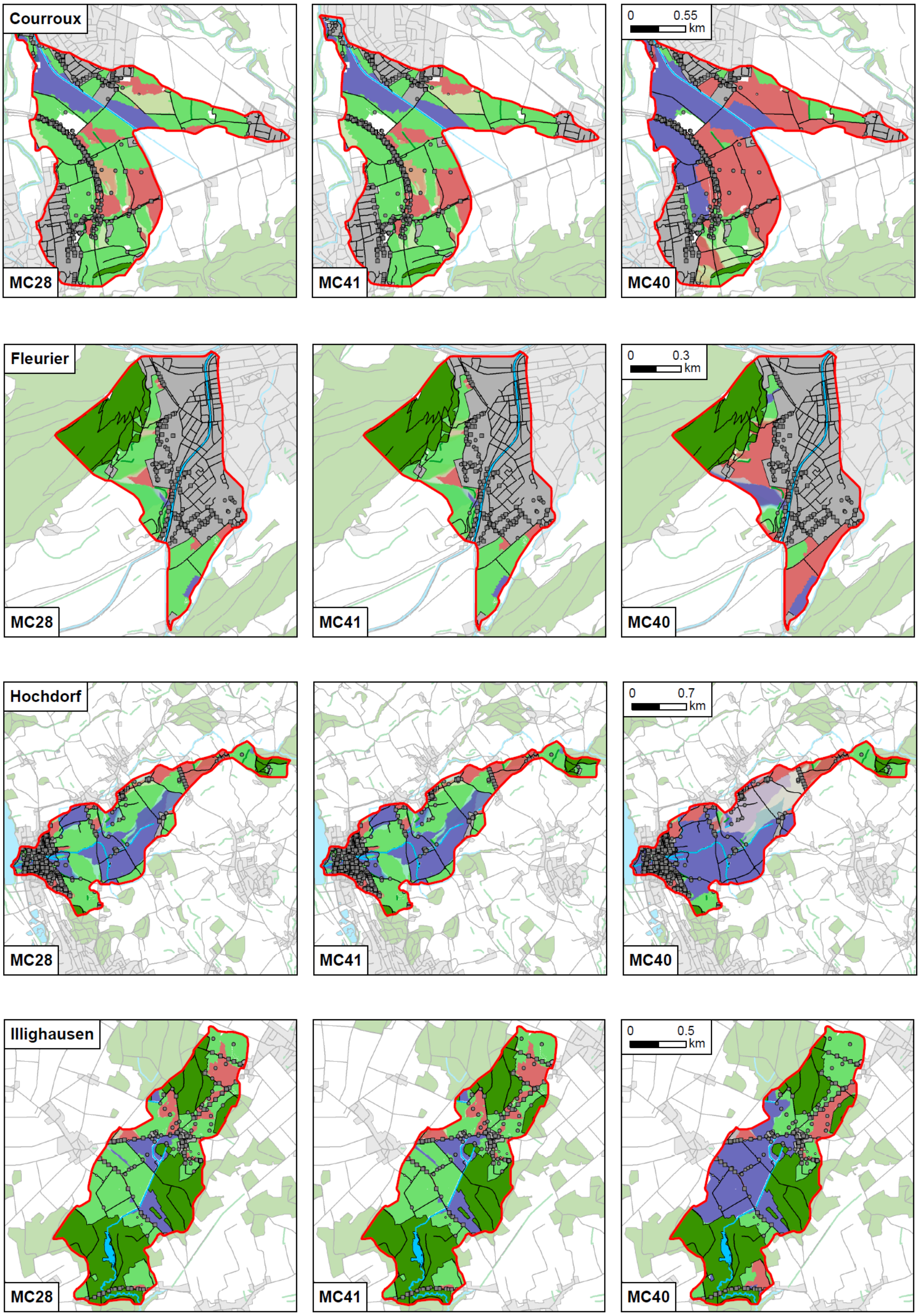
Figure S18: Fraction of maintenance shafts per study area belonging to a certain landscape element

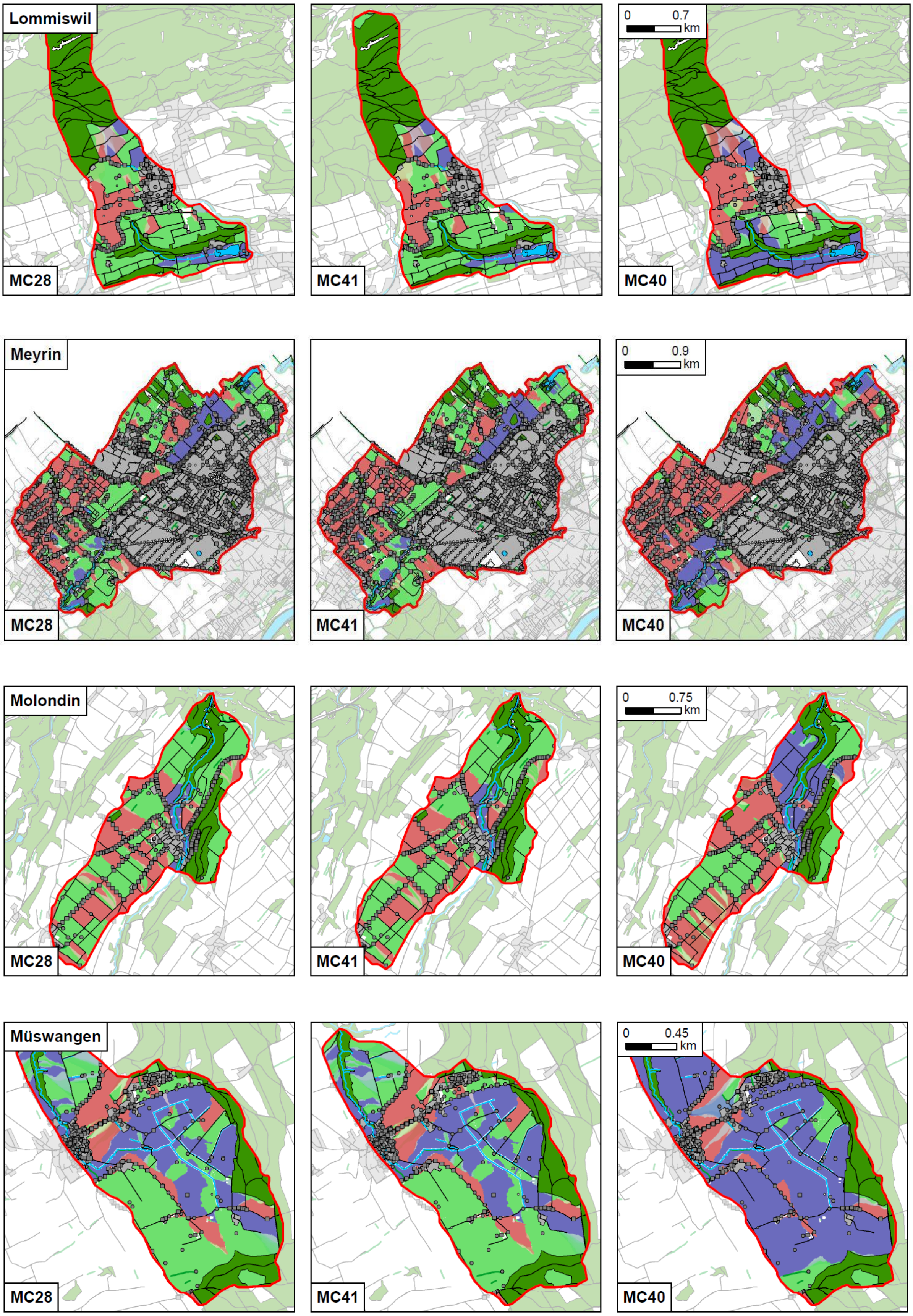
S2.2.2 Surface runoff connectivity: Study areas

S2.2.2.1 Example results for each study area

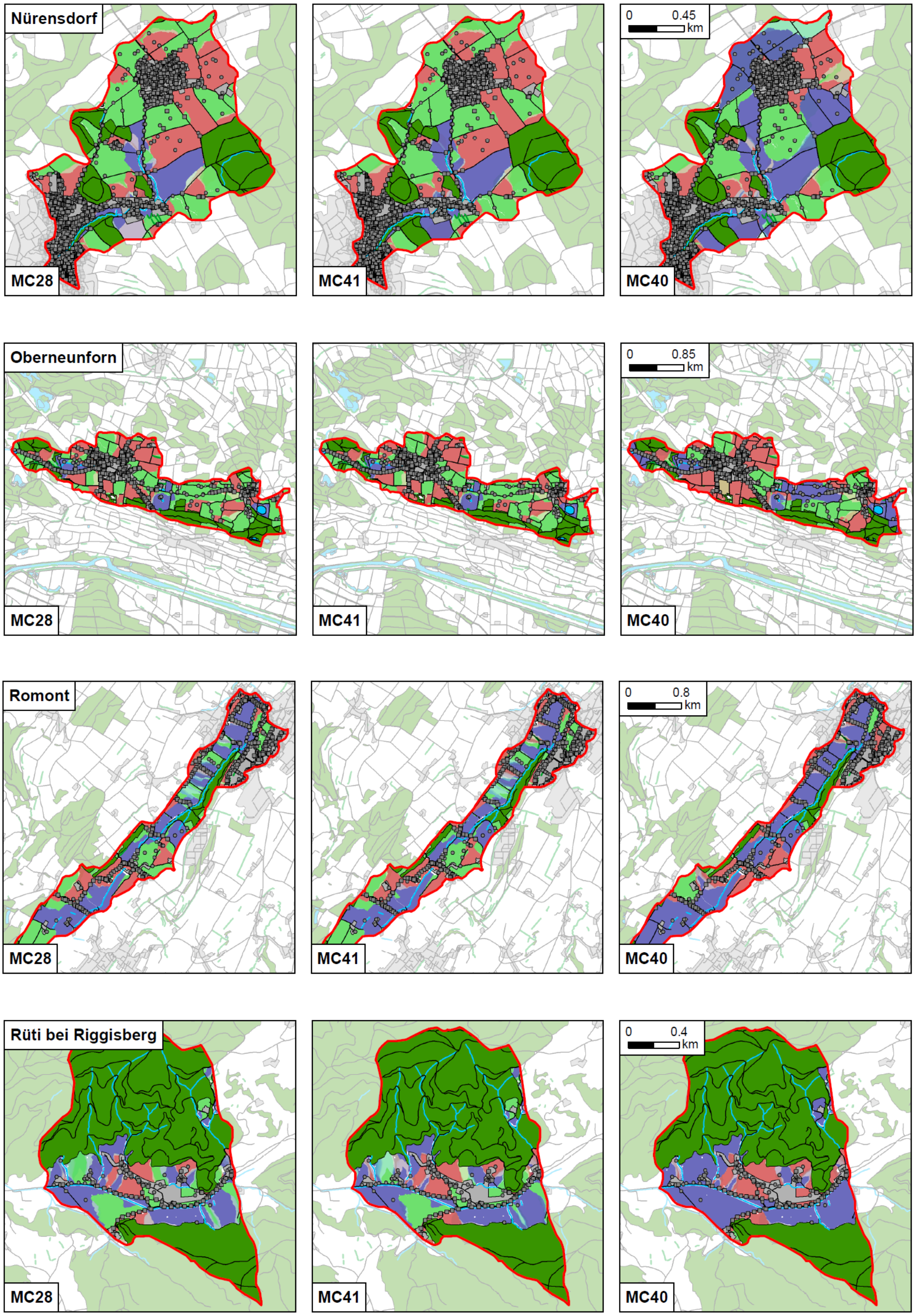
In the following, three example Monte Carlo analysis results (MC28, MC41, and MC40) are given for each of the study areas. The figures below correspond to Figure 7 in the main part of the article.

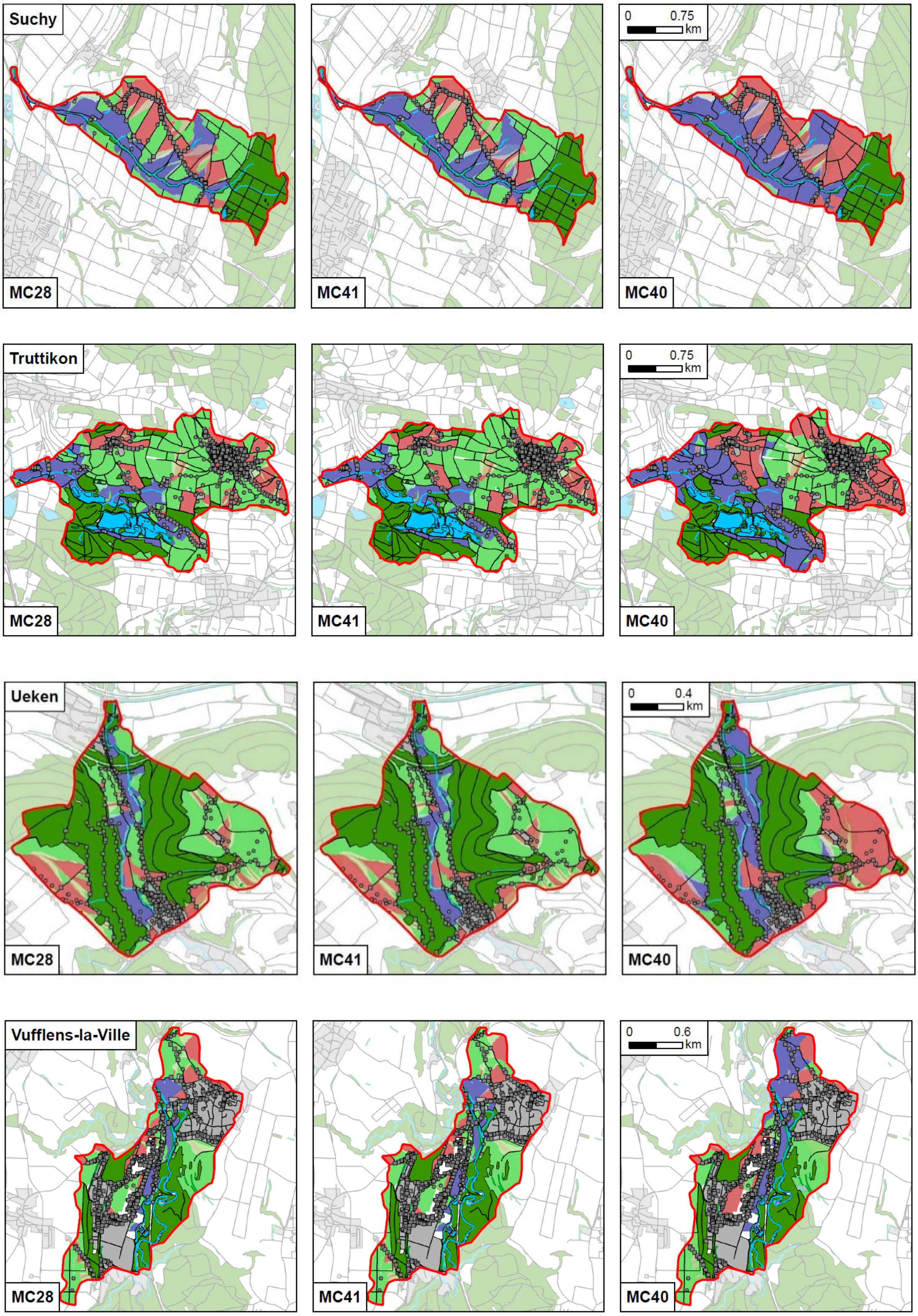



Courroux
MC28
MC41
0 0.55 km
MC40
Fleurier
MC28
MC41
0 0.3 km
MC40
Hochdorf
MC28
MC41
0 0.7 km
MC40
Illighausen
MC28
MC41
0 0.5 km
MC40



Lommiswil
MC28
MC41
0 0.7 km
MC40
Meyrin
MC28
MC41
0 0.9 km
MC40
Molondin
MC28
MC41
0 0.75 km
MC40
Müswangen
MC28
MC41
0 0.45 km
MC40


Nürensdorf
MC28
MC41
0 0.45 km
MC40
Oberneunforn
MC28
MC41
0 0.85 km
MC40
Romont
MC28
MC41
0 0.8 km
MC40
Rüti bei Riggisberg
MC28
MC41
0 0.4 km
MC40


Suchy
MC28
MC41
0 0.75 km
MC40
Truttikon
MC28
MC41
0 0.75 km
MC40
Ueken
MC28
MC41
0 0.4 km
MC40
Vufflens-la-Ville
MC28
MC41
0 0.6 km
MC40

S2.2.2.2 Monte Carlo Results: Directly, indirectly, and not connected areas

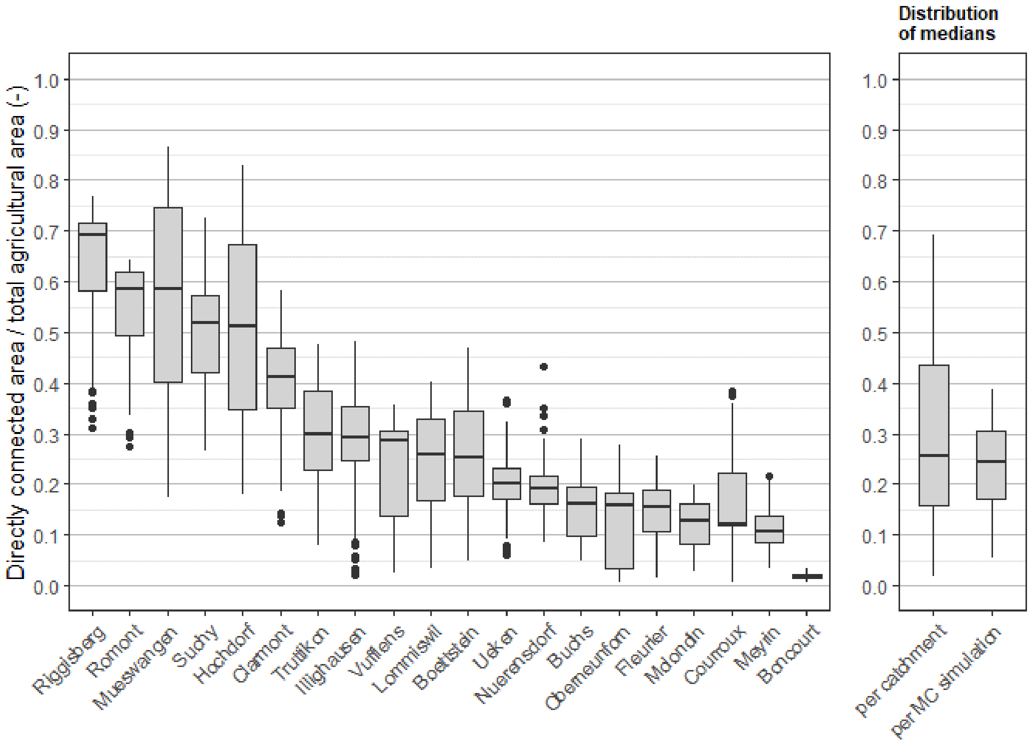


Figure S19: Left: Directly connected area per total agricultural area (-) as calculated by the Monte Carlo analysis for each study area. Right: Distribution of medians of directly connected area per total agricultural area (-) per study area and per Monte Carlo simulation.

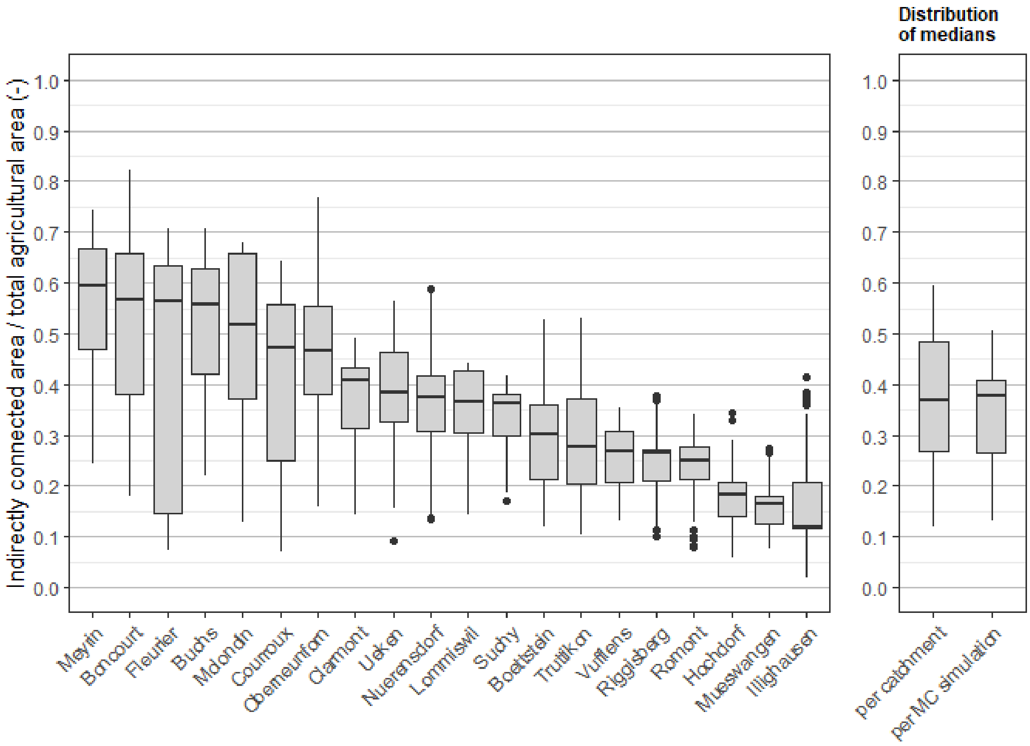


Figure S20: Indirectly connected area per total agricultural area (-) as calculated by the Monte Carlo analysis for each study area. Right: Distribution of medians of indirectly connected area per total agricultural area (-) per study area and per Monte Carlo simulation.

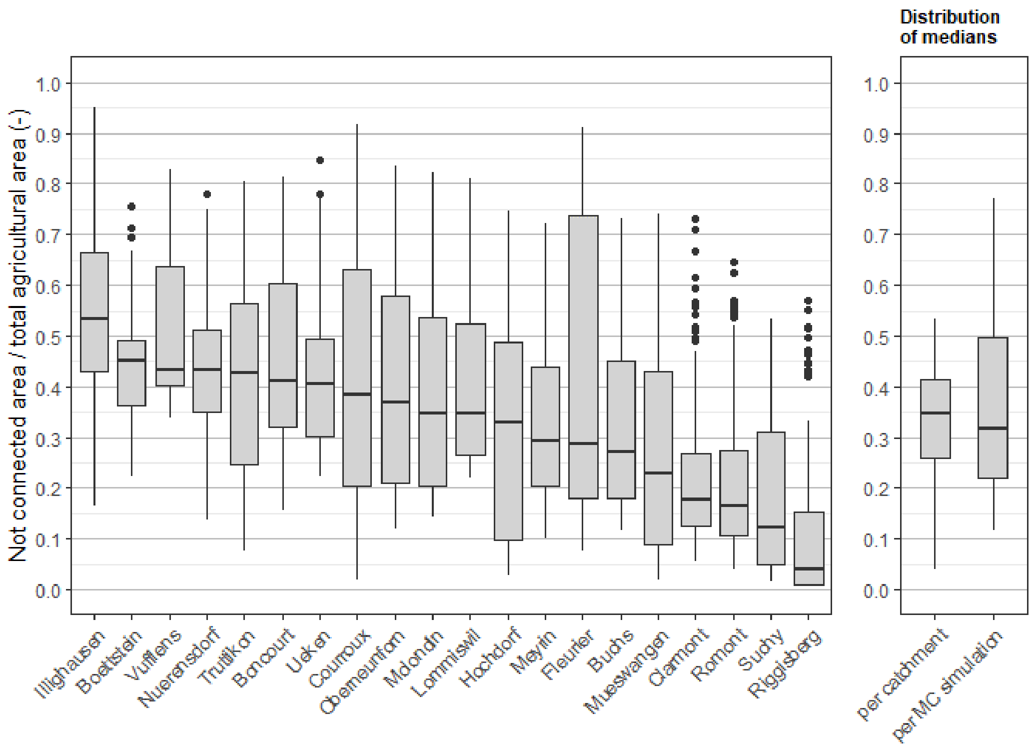


Figure S21: Not connected area per total agricultural area (-) as calculated by the Monte Carlo analysis for each study area. Right: Distribution of medians of not connected area per total agricultural area (-) per study area and per Monte Carlo simulation.

S2.2.2.3 Correlation of connectivity fractions with catchment statistics

Table S8: Correlation of catchment statistics with fractions of connected area connectivity. NECM: National erosion connectivity model, LSCM: Local surface runoff connectivity model.

Variable	Fraction directly connected $f_{LSCM,dir}$ (-)			Fraction indirectly connected $f_{LSCM,indir}$ (-)			Fraction not connected $f_{LSCM,nc}$ (-)		
	R^2	Slope	p	R^2	Slope	p	R^2	Slope	p
NECM: Directly connected agricultural area per total agricultural area $f_{NECM,dir}$ (-)	0.71	1.0E+00	< 0.001 ***	-	-	-	-	-	-
NECM: Indirectly connected agricultural area per total agricultural area $f_{NECM,indir}$ (-)	-	-	-	0.52	6.0E-01	< 0.001 ***	-	-	-
NECM: Not connected agricultural area per total agricultural area $f_{NECM,nc}$ (-)	-	-	-	-	-	-	0.26	4.0E-01	0.022 *
Surface water body density (m^{-1})	0.51	2.2E+02	< 0.001 ***	0.35	-1.4E+02	0.006 **	0.14	-7.6E+01	0.10 *
Paved road density (m^{-1})	0.20	-2.2E+01	0.049 *	0.19	1.7E+01	0.053 -	0.04	6.5E+00	0.41 -
Inlet shaft density (ha^{-1})	0.07	-1.3E-01	0.28 -	0.10	1.2E-01	0.17 -	0.00	1.0E-02	0.90 -
Maintenance shaft density (ha^{-1})	0.15	4.0E+02	0.09 -	0.07	-2.0E+02	0.27 -	0.07	-1.8E+02	0.27 -
Yearly rainfall (mm/year)	0.10	-5.2E-02	0.17 -	0.06	3.2E-02	0.28 -	0.04	2.0E-02	0.43 -
Total road density (m^{-1})	0.05	2.6E-01	0.35 -	0.05	-2.0E-01	0.33 -	0.00	-4.5E-02	0.80 -
Subsurface waterbody density (m^{-1})	0.11	-7.5E+00	0.14 -	0.04	3.3E+00	0.40 -	0.10	4.5E+00	0.18 -
Fraction of agricultural area (-)	0.00	2.6E+01	0.94 -	0.03	-1.7E+02	0.48 -	0.03	1.7E+02	0.43 -
Unpaved road density (m^{-1})	0.15	4.4E-04	0.09 -	0.02	-1.2E-04	0.55 -	0.18	-3.2E-04	0.063 -
Lake shore density (m^{-1})	0.03	1.3E-02	0.49 -	0.02	7.7E-03	0.60 -	0.13	-1.9E-02	0.13 -
Slope on agricultural areas (°)	0.04	-5.8E+00	0.41 -	0.00	2.2E-01	0.97 -	0.09	6.0E+00	0.19 -

S2.2.2.4 Sensitivity analysis

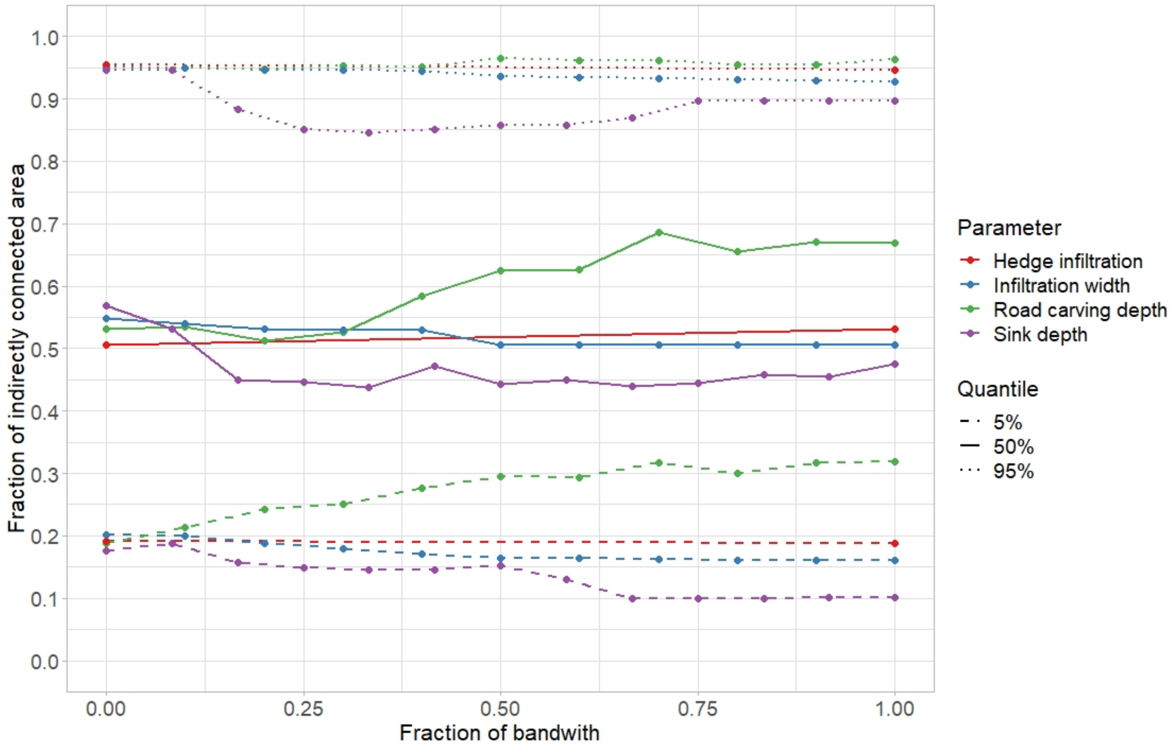


Figure S22: Sensitivity analysis for shortcut definition A. The y-axis shows the fraction of indirectly connected area per total connected area. The parameters were varied within the following bandwidths: Hedge infiltration [no; yes], infiltration width [6 m; 100 m], road carving depth [0 cm; 100 cm], sink depth [0 cm; 100 cm]

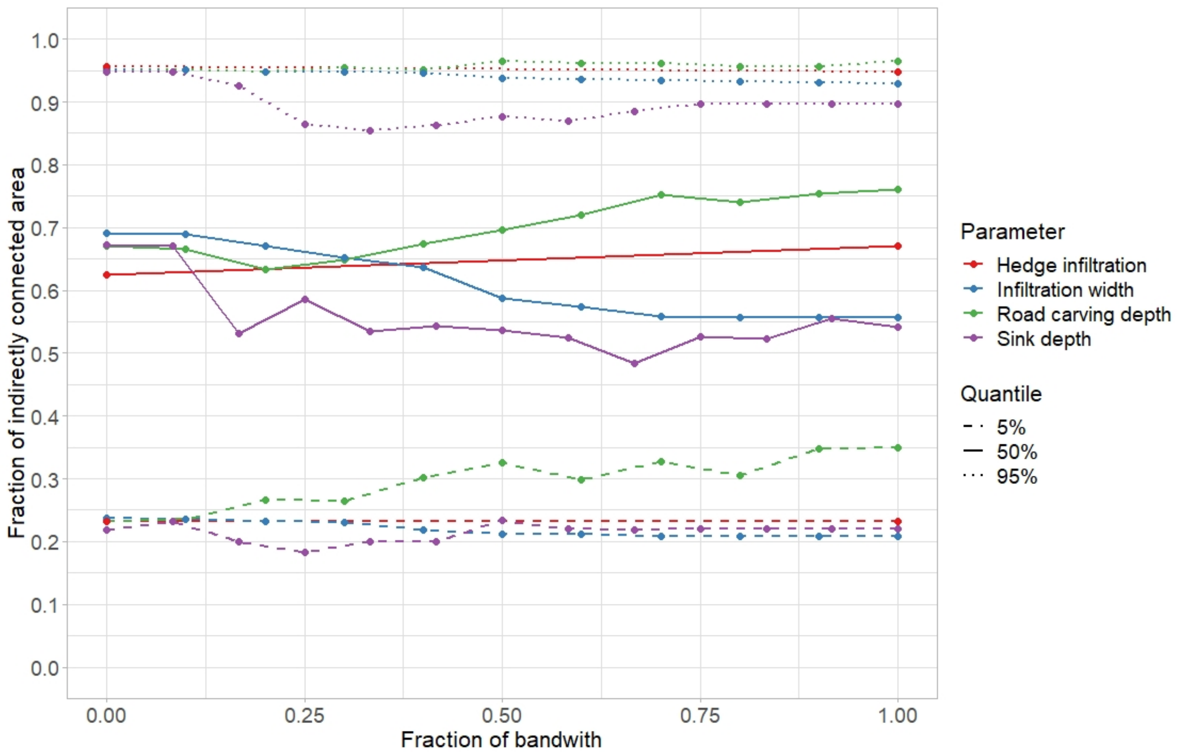


Figure S23: Sensitivity analysis for shortcut definition B. The y-axis shows the fraction of indirectly connected area per total connected area. The parameters were varied within the following bandwidths: Hedge infiltration [no; yes], infiltration width [6 m; 100 m], road carving depth [0 cm; 100 cm], sink depth [0 cm; 100 cm]

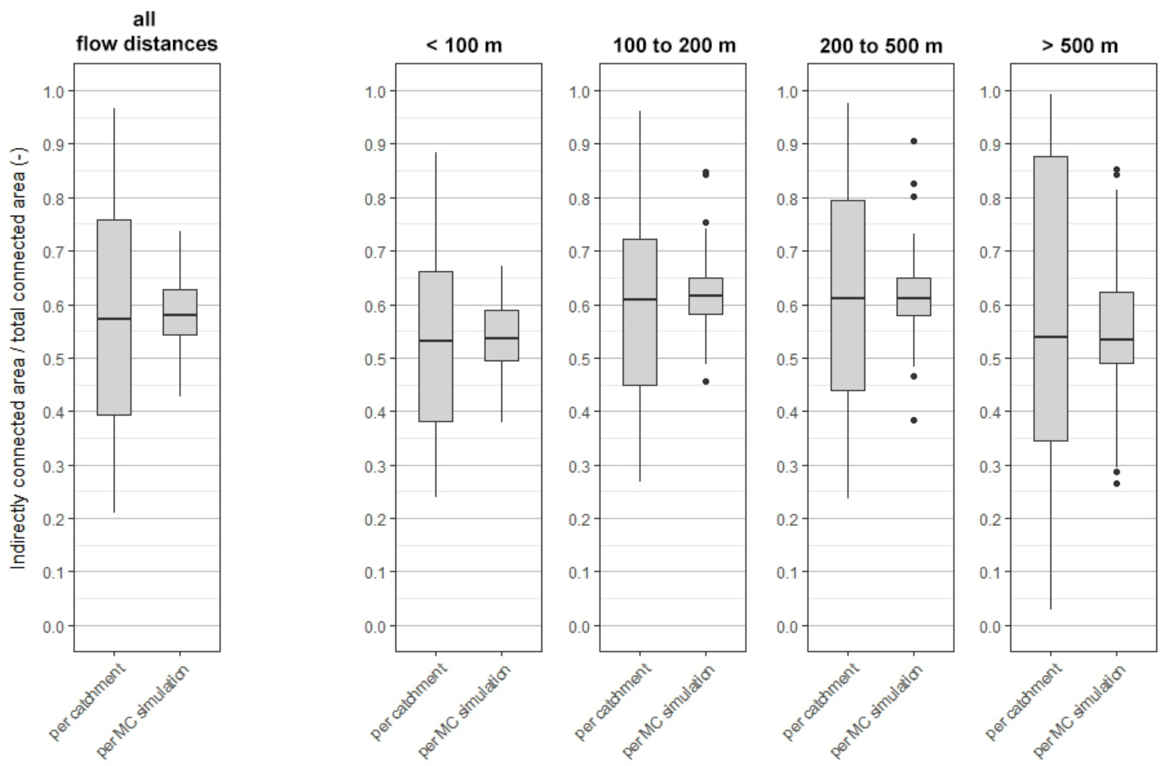


Figure S24: Influence of flow distance on Monte Carlo results. Distribution of medians of indirectly connected area per total connected area (-) per study area and per Monte Carlo simulation for different flow distances. Left: Consideration of all flow distances. Right: Consideration of flow distances of smaller than 100 m, 100 to 200 m, 200 to 500 m, and larger than 500 m, respectively.

S2.2.2.5 Distribution of slope and wetness index

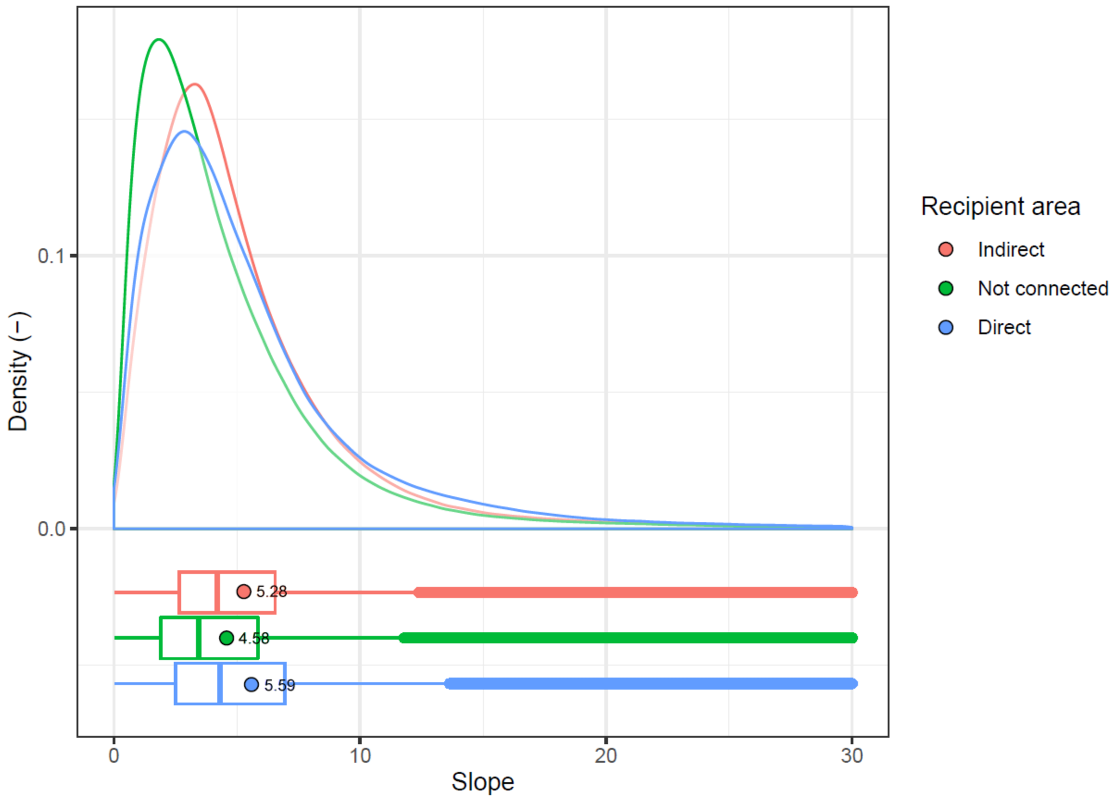


Figure S25: Slope distribution (degrees) on different source area types

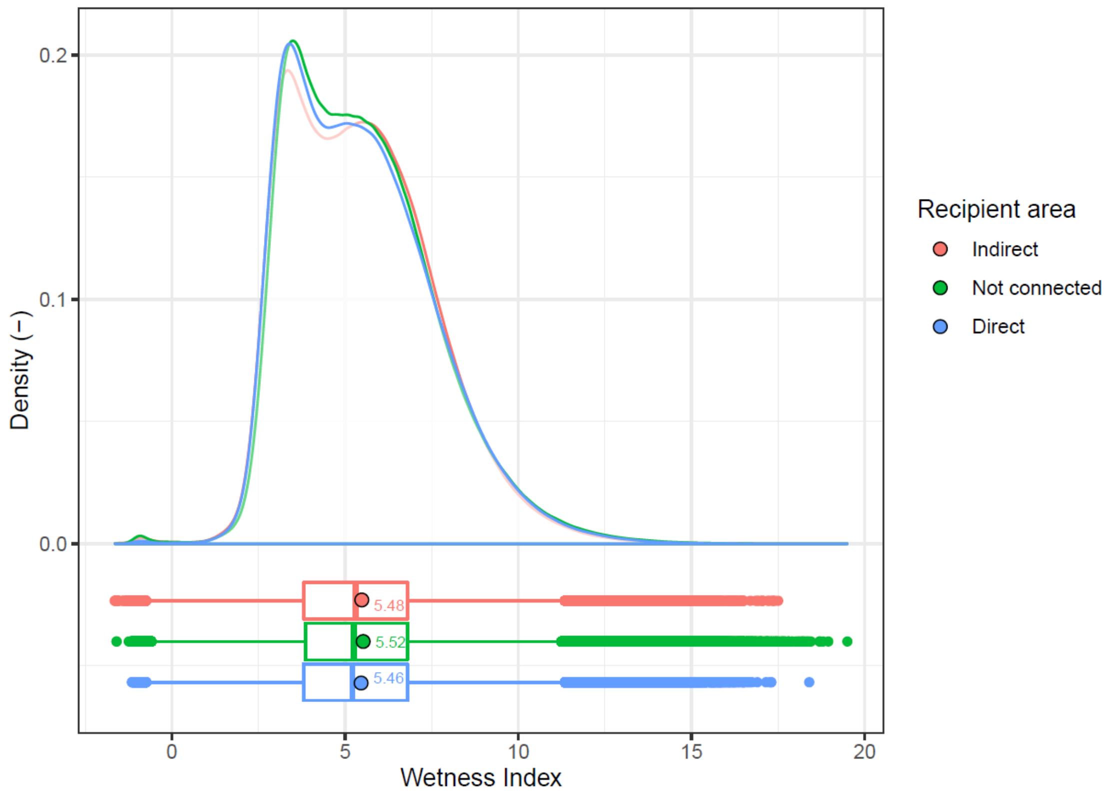


Figure S26: Topographic wetness index distribution (-) on different source area types

S2.2.3 Surface runoff connectivity: Extrapolation to national level

S2.2.3.1 National area fractions

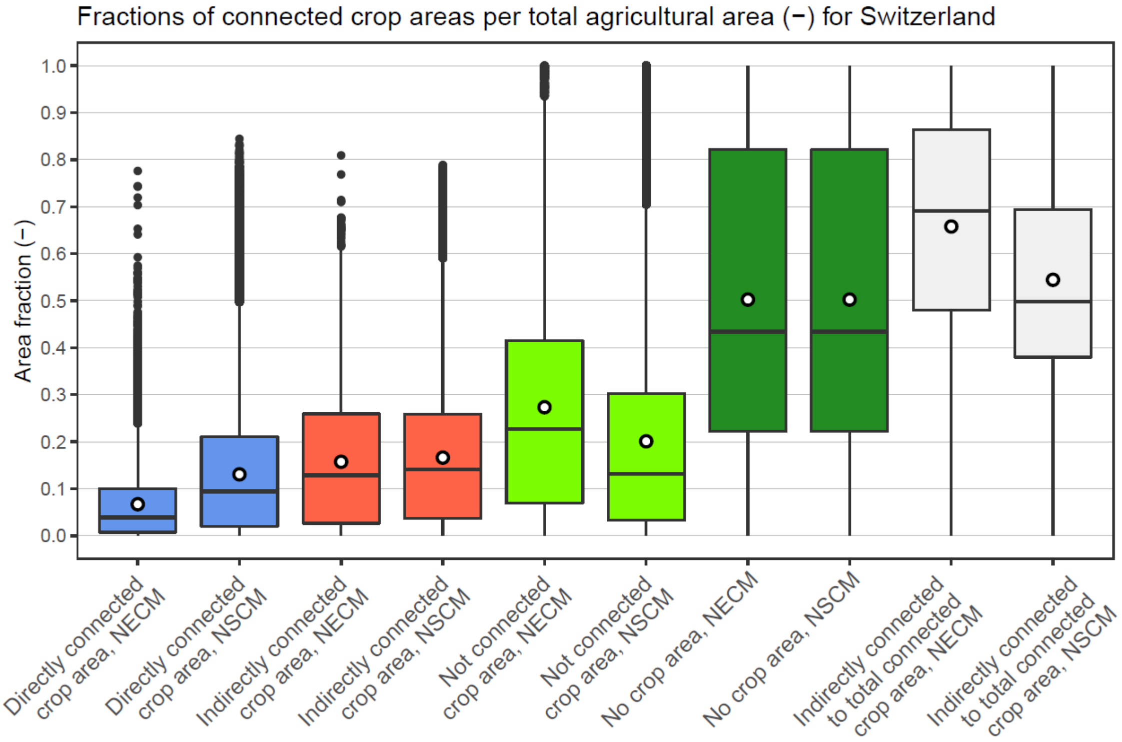


Figure S27: Modelled area fractions by the NECM and the NSCM: Directly, indirectly, and not connected crop areas per total agricultural area, non-cropping area per total agricultural area, and indirectly connected crop area per total connected crop area for all catchments in Switzerland.

Table S9: Statistics of modelled area fraction by the NECM and the NSCM. For the NSCM, the mean, the 5% quantile and the 95% quantile of the mean fractions resulting from the MC simulations is given. Additionally, the mean, the 5% quantile and the 95% quantile of the mean fractions resulting from the bootstrapping approach is given.

Statistic	**Fraction of directly connected crop area $f_{crop,dir}$**	**Fraction of indirectly connected crop area $f_{crop,indir}$**	**Fraction of not connected crop area $f_{crop,nc}$**	**No crop area**	**Fraction of indirectly per total connected area $f_{fracindir}$**
NECM	6.7%	16%	27%	50%	66%
NSCM: Mean (5% quantile; 95% quantile) of mean per MC simulation	13% (6.9%; 18%)	17% (7.0%; 24%)	20% (8.8%; 36%)	50% (50%; 50%)	54% (47%; 60%)
NSCM: Mean (5% quantile; 95% quantile) of mean per bootstrap simulation	14% (11%; 16%)	15% (13%; 17%)	21% (19%; 24%)	50% (50%; 50%)	49% (42%; 55%)

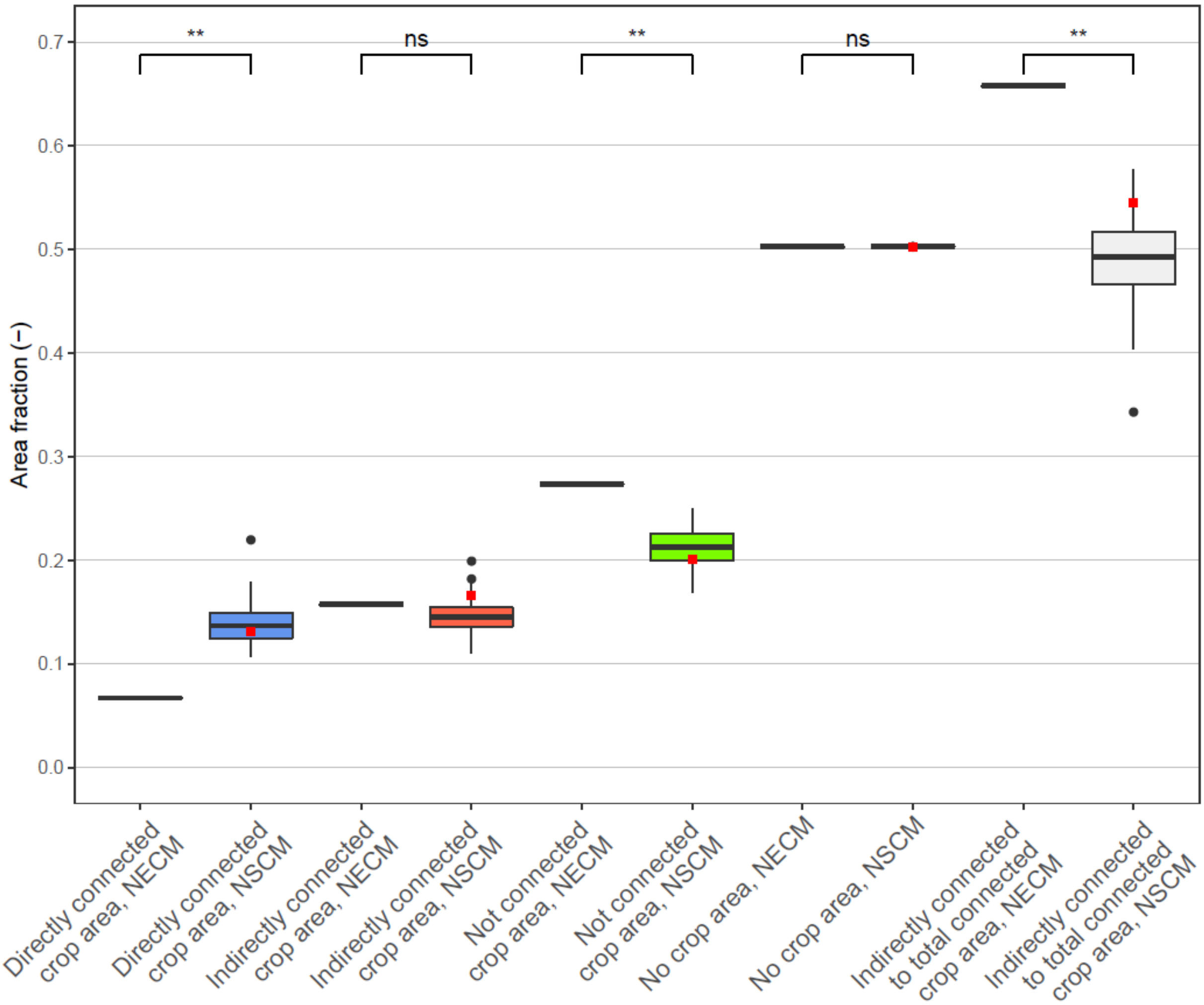


Figure S28: Mean area fractions reported by the NECM and distribution of the bootstrapped mean area fractions reported by the NSCM. Directly, indirectly, and not connected crop areas per total agricultural area, non-cropping area per total agricultural area, and indirectly connected crop area per total connected crop area for all catchments in Switzerland. The red squares report the means reported by the NSCM without using a bootstrapping approach. The black lines on the top of the plot indicate if the mean fraction reported by the NECM is significantly different from the distribution of means reported by the bootstrapping approach (: p < 0.01, ns: not significant). Significance values were determined from the empirical cumulative distribution of the bootstrapped means.**

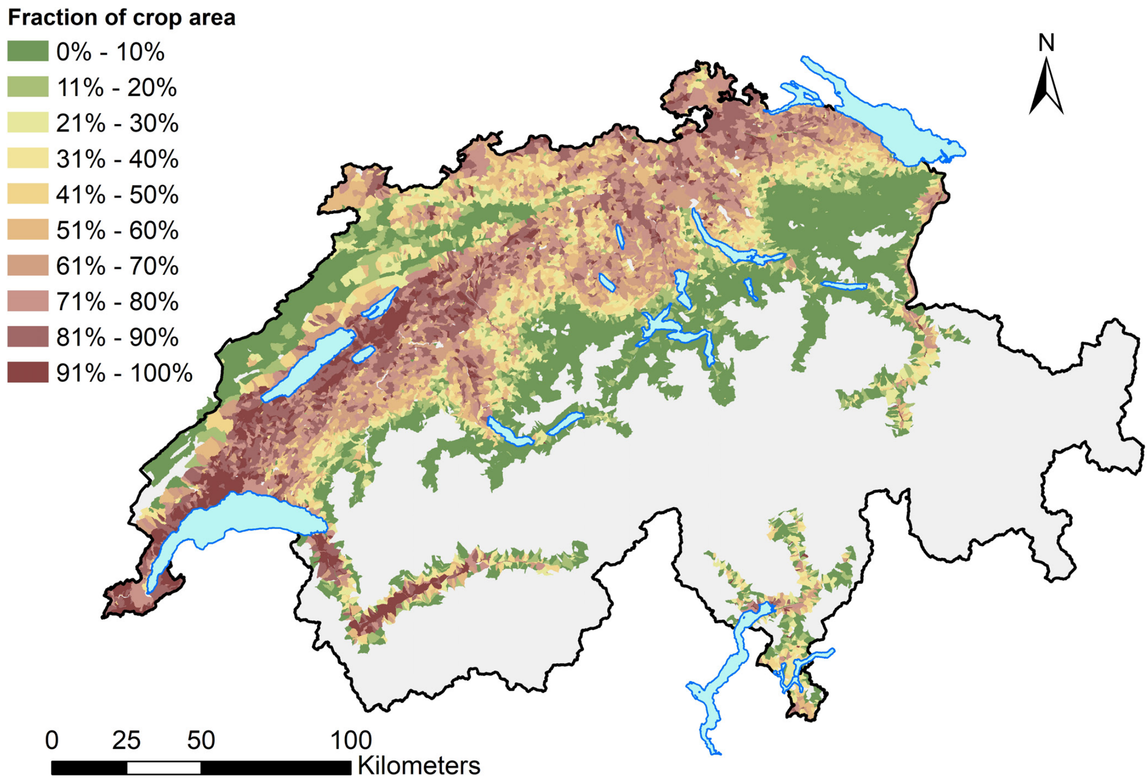


Figure S29: Fraction of crop area (arable land, vineyards, orchards, horticulture) per total agricultural area per catchment. Source of background map: Swisstopo (2010)

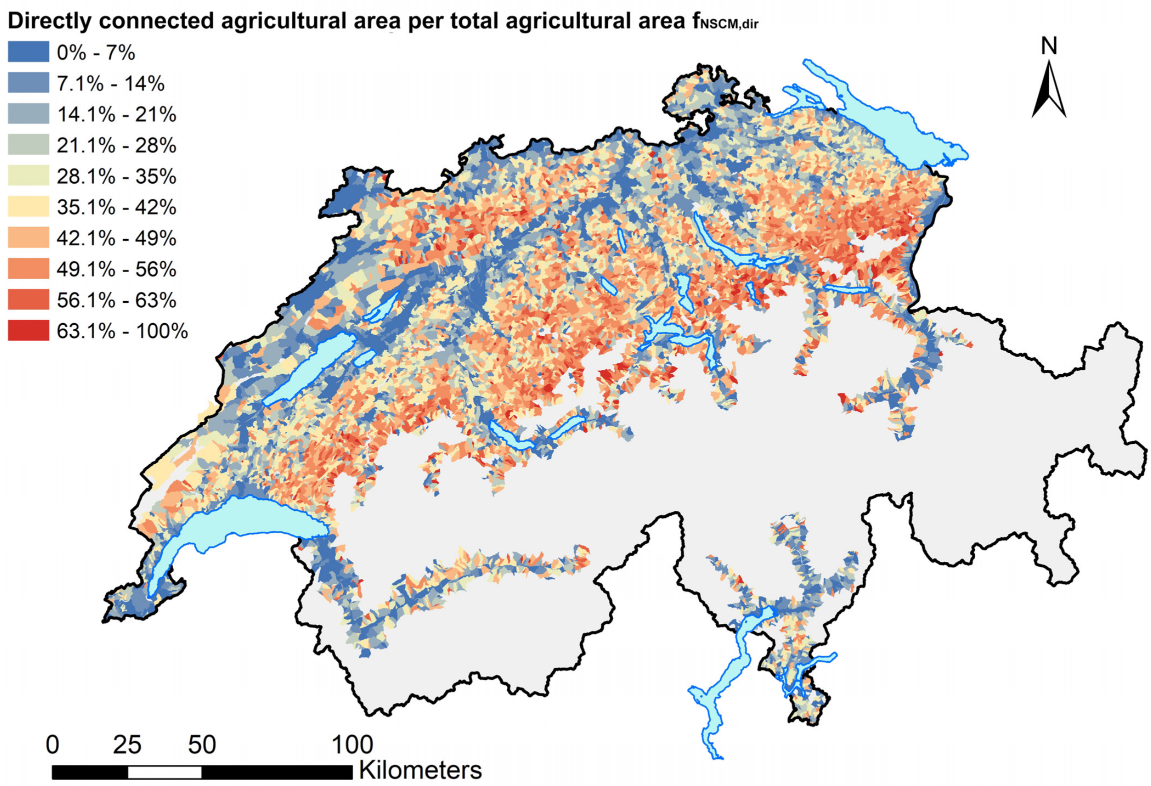


Figure S30: Fraction of directly connected agricultural area per total agricultural area per catchment $f_{NSCM,dir}$. Source of background map: Swisstopo (2010)

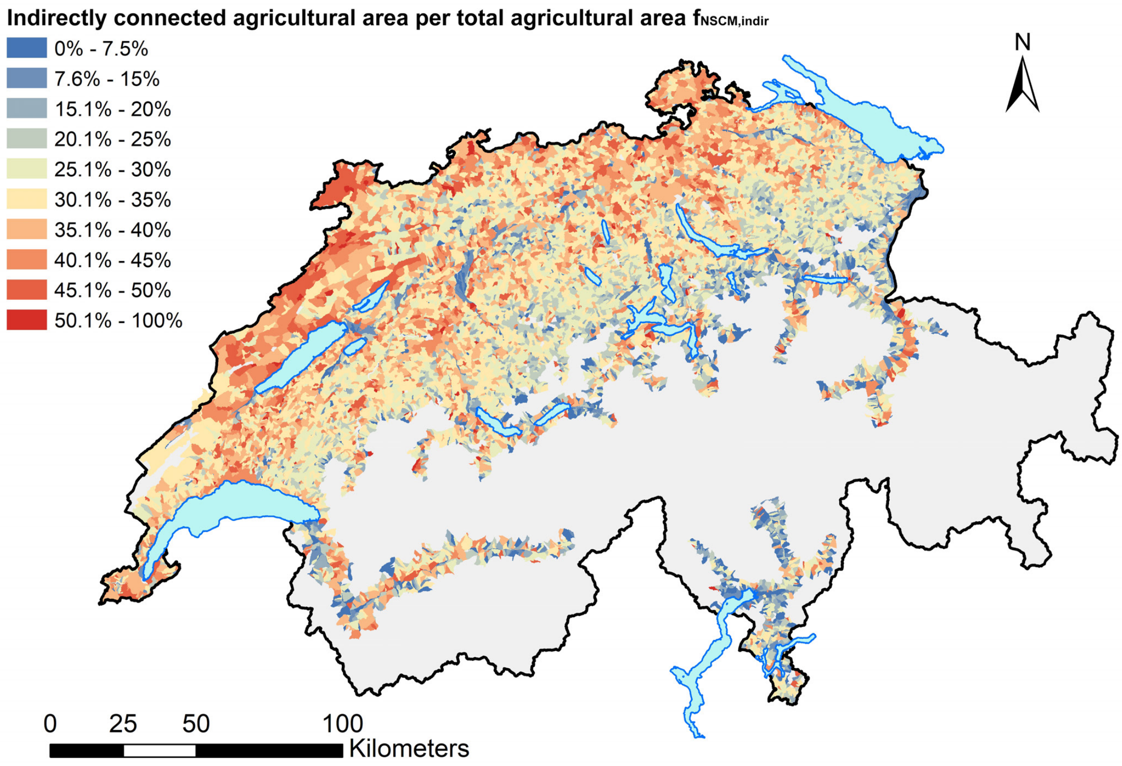


Figure S31: Fraction of indirectly connected agricultural area per total agricultural area per catchment $f_{NSCM,indir}$. Source of background map: Swisstopo (2010)

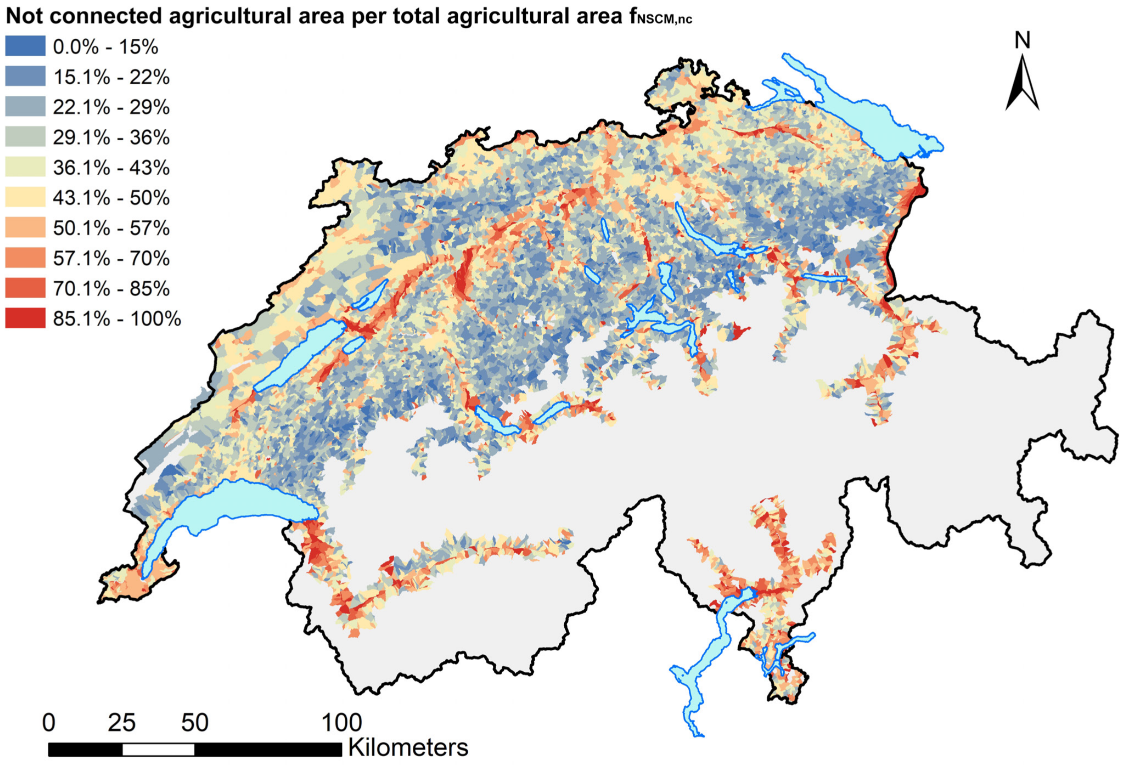


Figure S32: Fraction of not connected agricultural area per total agricultural area per catchment $f_{NSCM,nc}$. Source of background map: Swisstopo (2010)

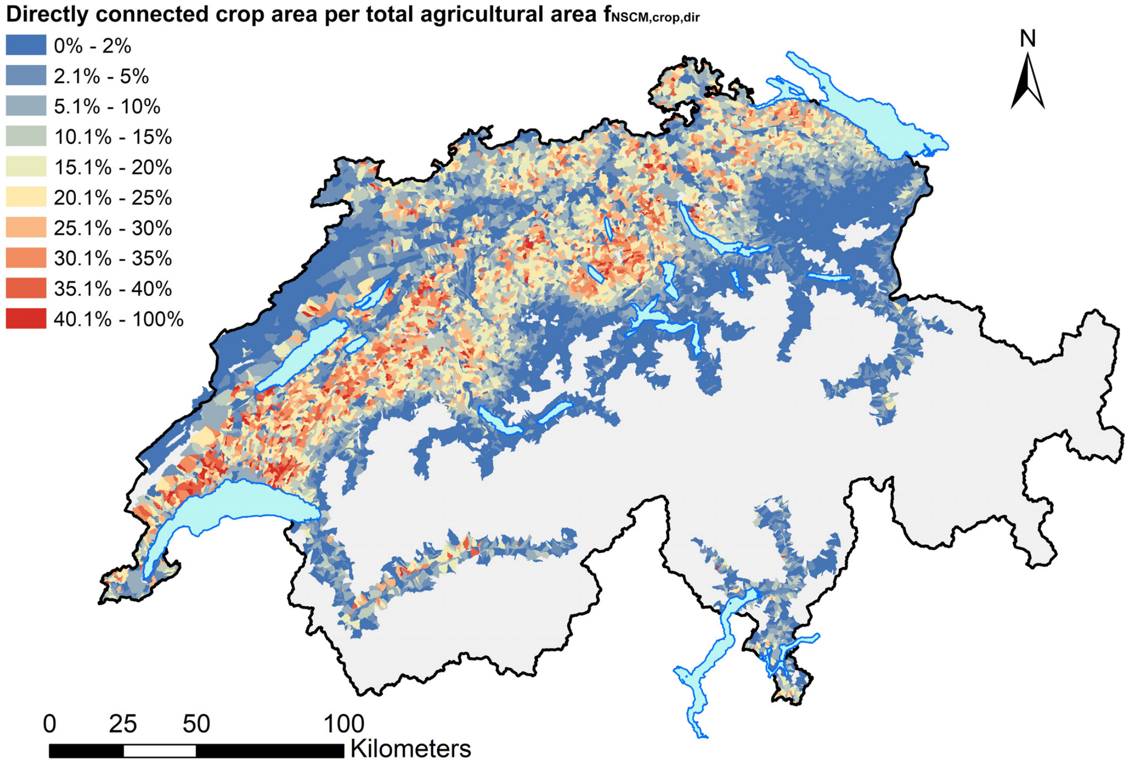


Figure S33: Fraction of directly connected crop area per total agricultural are per catchment $f_{NSCM,crop,dir}$. Source of background map: Swisstopo (2010)

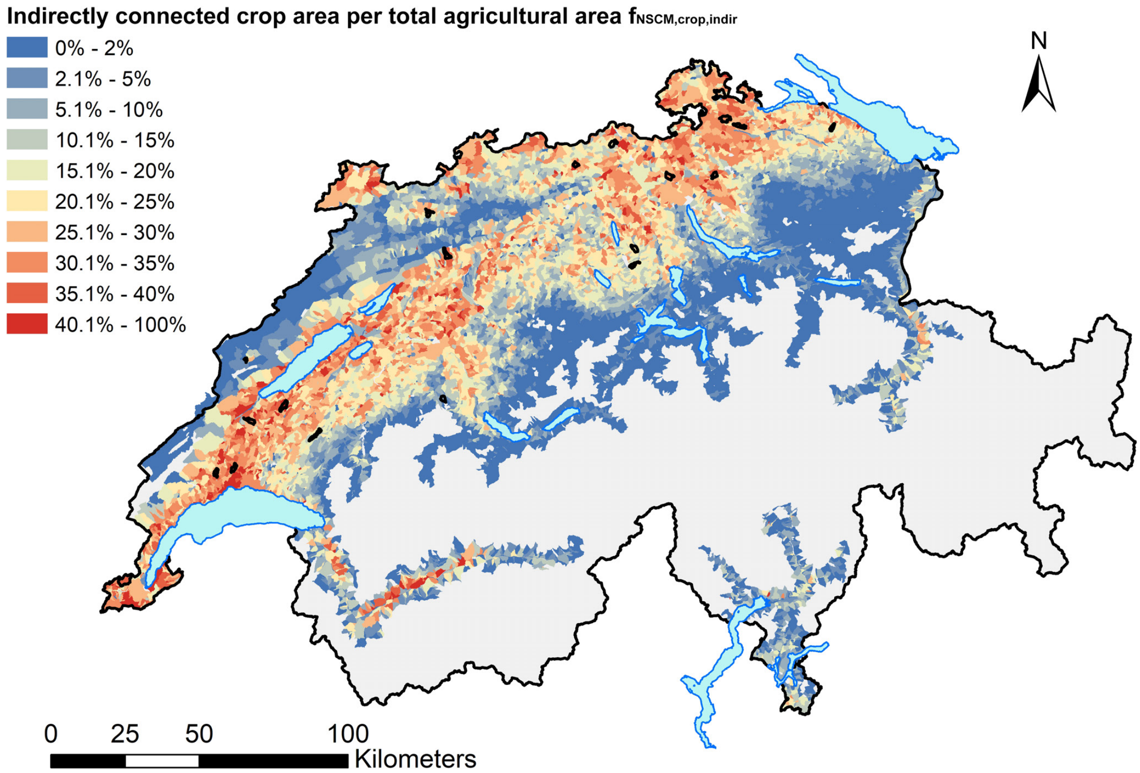


Figure S34: Fraction of indirectly connected crop area per total agricultural are per catchment $f_{NSCM,crop,dir}$. Source of background map: Swisstopo (2010)

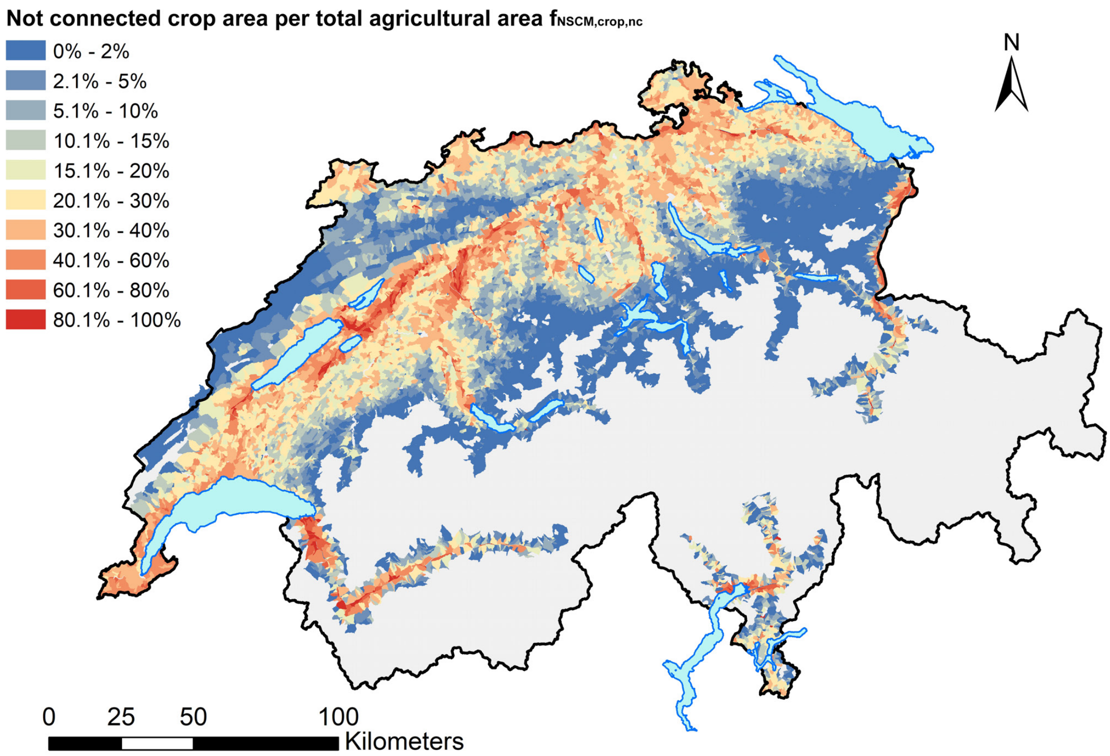


Figure S35: Fraction of not connected crop area per total agricultural area per catchment $f_{NSCM,crop,nc}$. Source of background map: Swisstopo (2010)

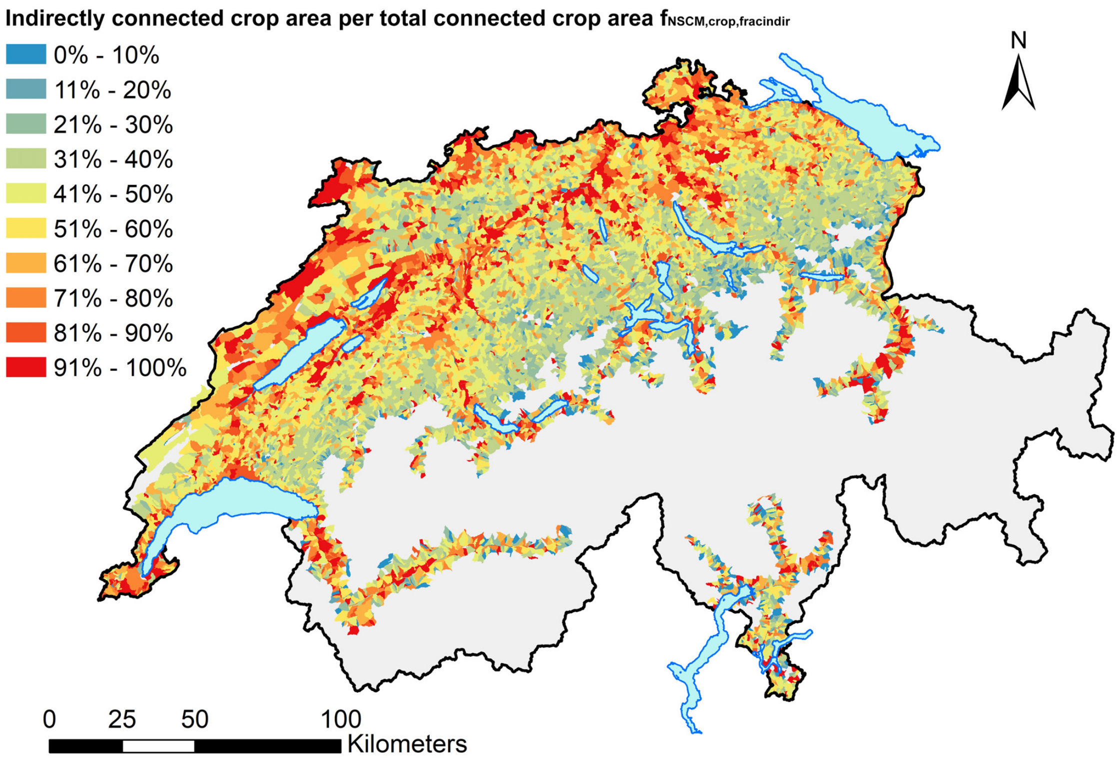


Figure S36: Fraction of indirectly connected crop area per total connected crop area $f_{NSCM,drop,fracindir}$. Source of background map: Swisstopo (2010)

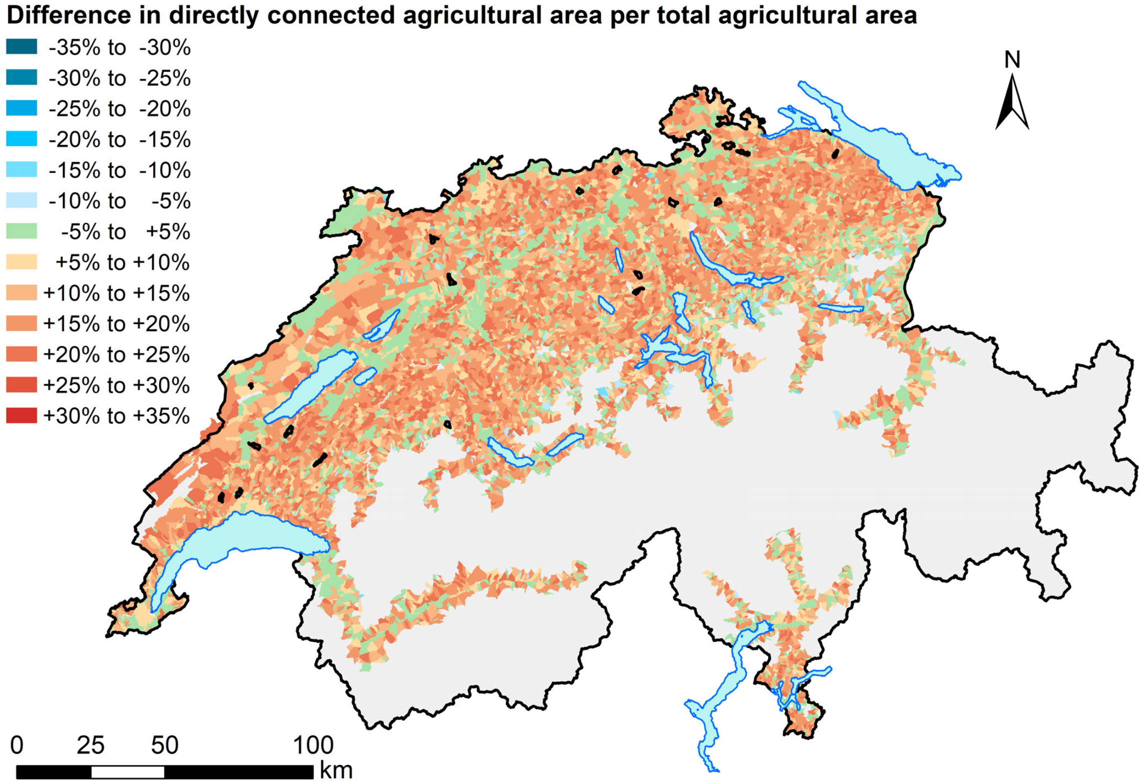


Figure S37: Difference between the fractions of directly connected agricultural area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,dir}$ - $f_{NECM,dir}$). Source of background map: Swisstopo (2010)

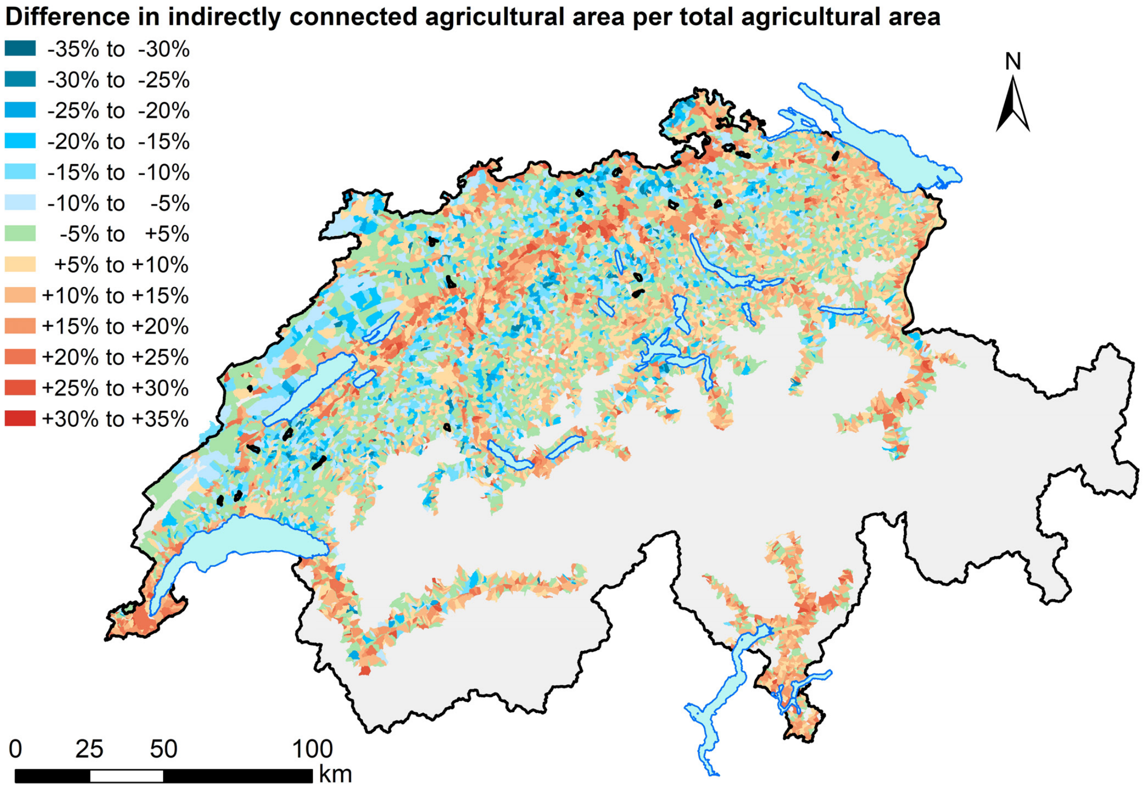


Figure S38: Difference between the fractions of indirectly connected agricultural area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,indir}$ - $f_{NECM,indir}$). Source of background map: Swisstopo (2010)

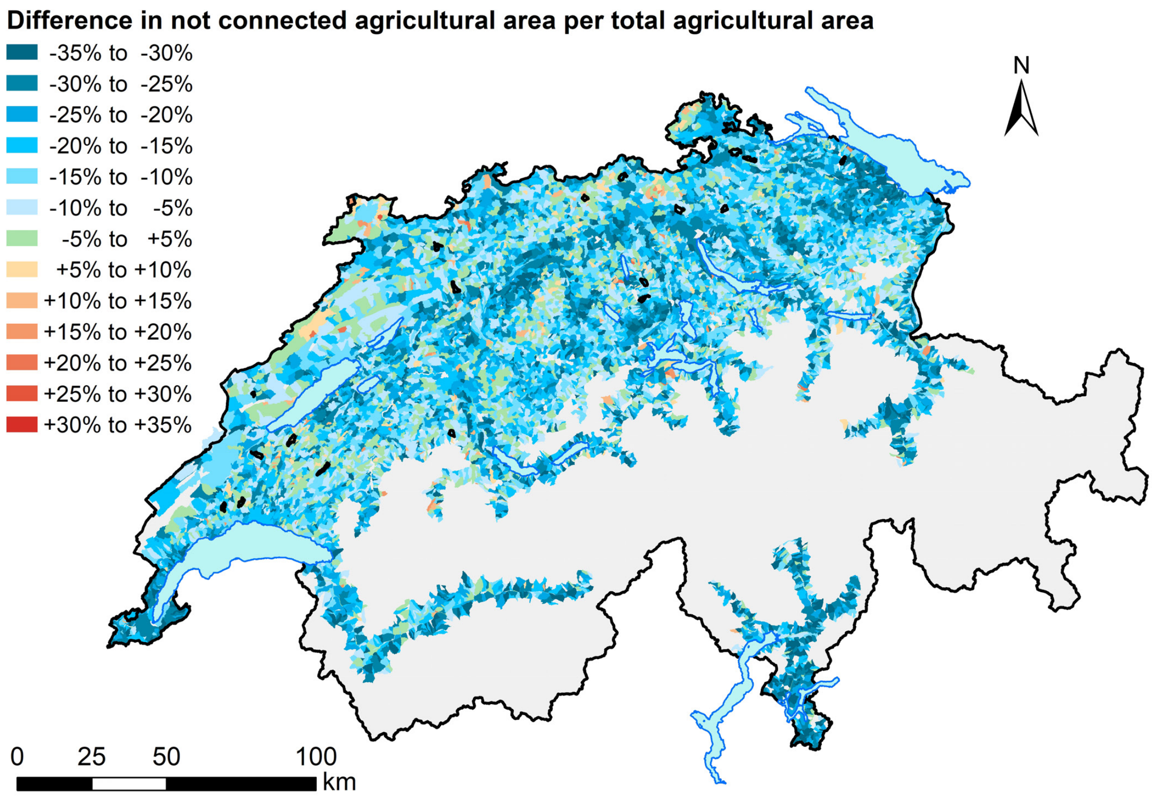


Figure S39: Difference between the fractions of not connected agricultural area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,nc}$ - $f_{NECM,nc}$). Source of background map: Swisstopo (2010)

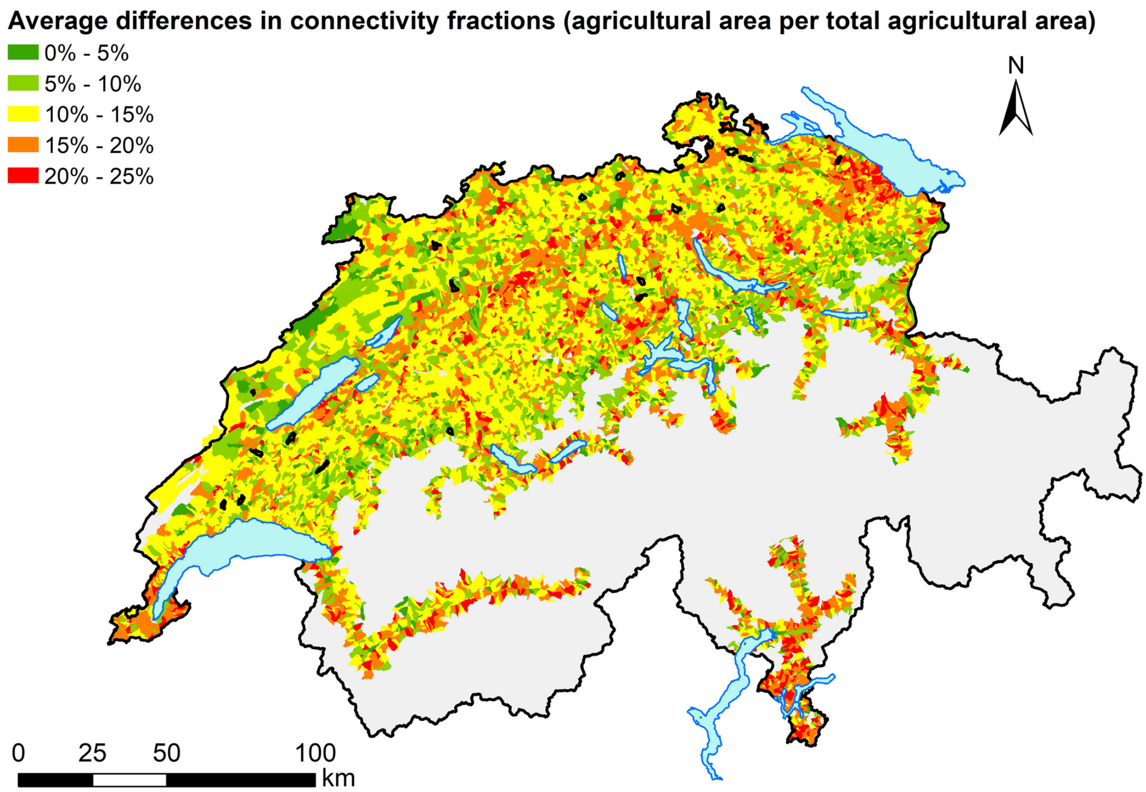


Figure S40: Average difference in connectivity fractions of agricultural areas reported by the NSCM and the NECM: Δf_{crop} = (($f_{NSCM,dir}$ - $f_{NECM,dir}$) + ($f_{NSCM,indir}$ - $f_{NECM,indir}$) + ($f_{NSCM,nc}$ - $f_{NECM,nc}$))/3. The map shows data for all Swiss catchments in the valley zones, hill zones and lower elevation mountain zones. Grey areas represent higher elevation mountain zones that were excluded from the analysis. Study areas are marked with black lines. Source of background map: Swisstopo (2010)

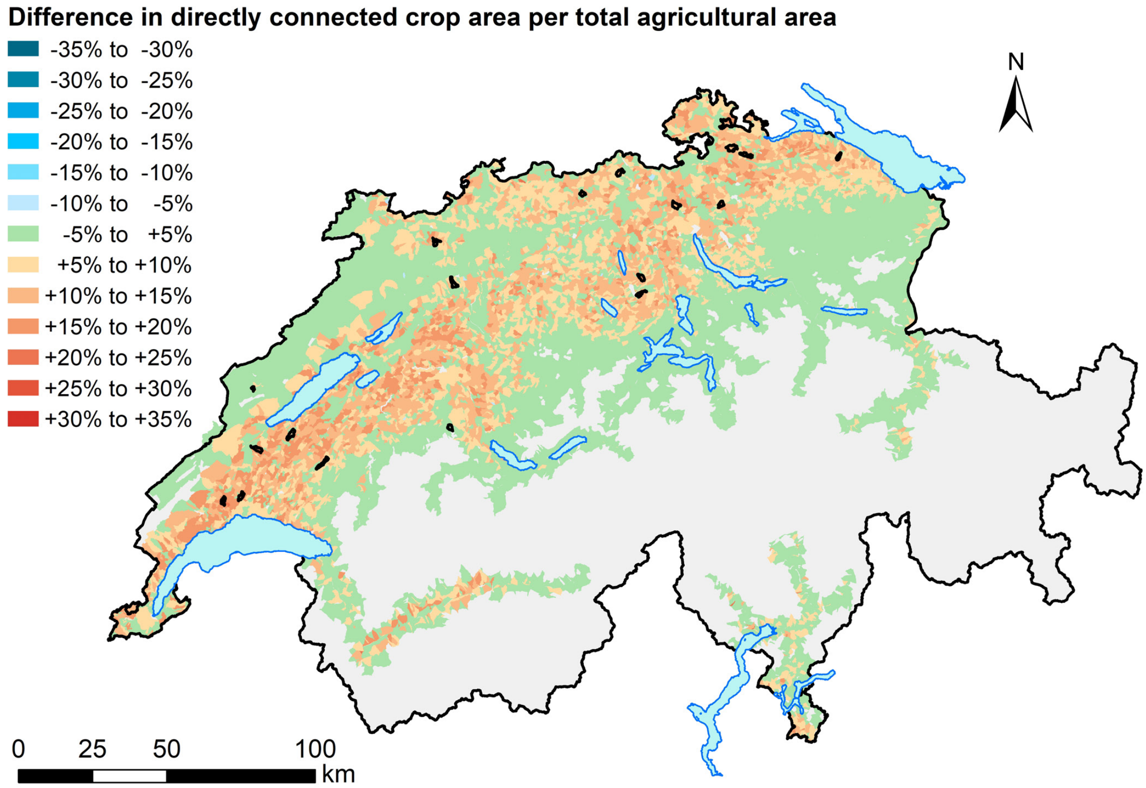


Figure S41: Difference between the fractions of directly connected crop area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,crop,dir}$ - $f_{NECM,crop,dir}$). Source of background map: Swisstopo (2010)

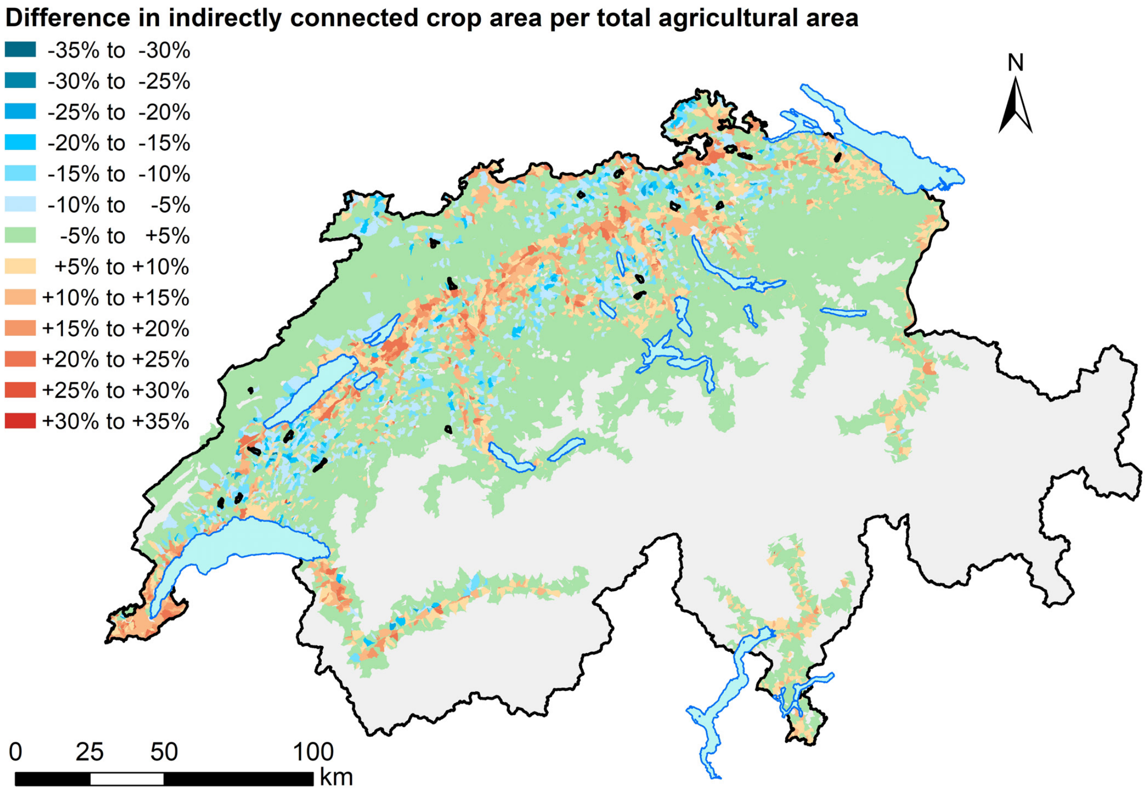


Figure S42: Difference between the fractions of indirectly connected crop area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,crop,indir}$ - $f_{NECM,crop,indir}$). Source of background map: Swisstopo (2010)

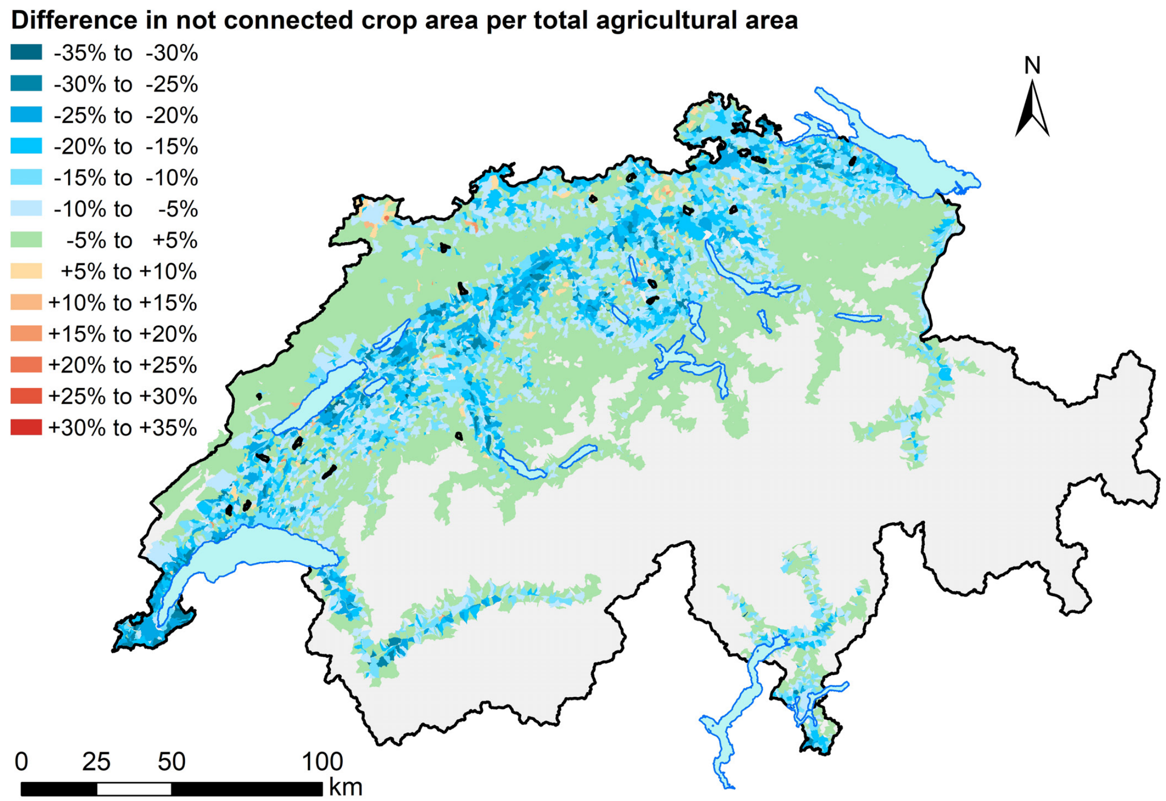


Figure S43: Difference between the fractions of not connected crop area per total agricultural area reported by the NSCM and the NECM ($f_{NSCM,crop,nc}$ - $f_{NECM,crop,nc}$). Source of background map: Swisstopo (2010)

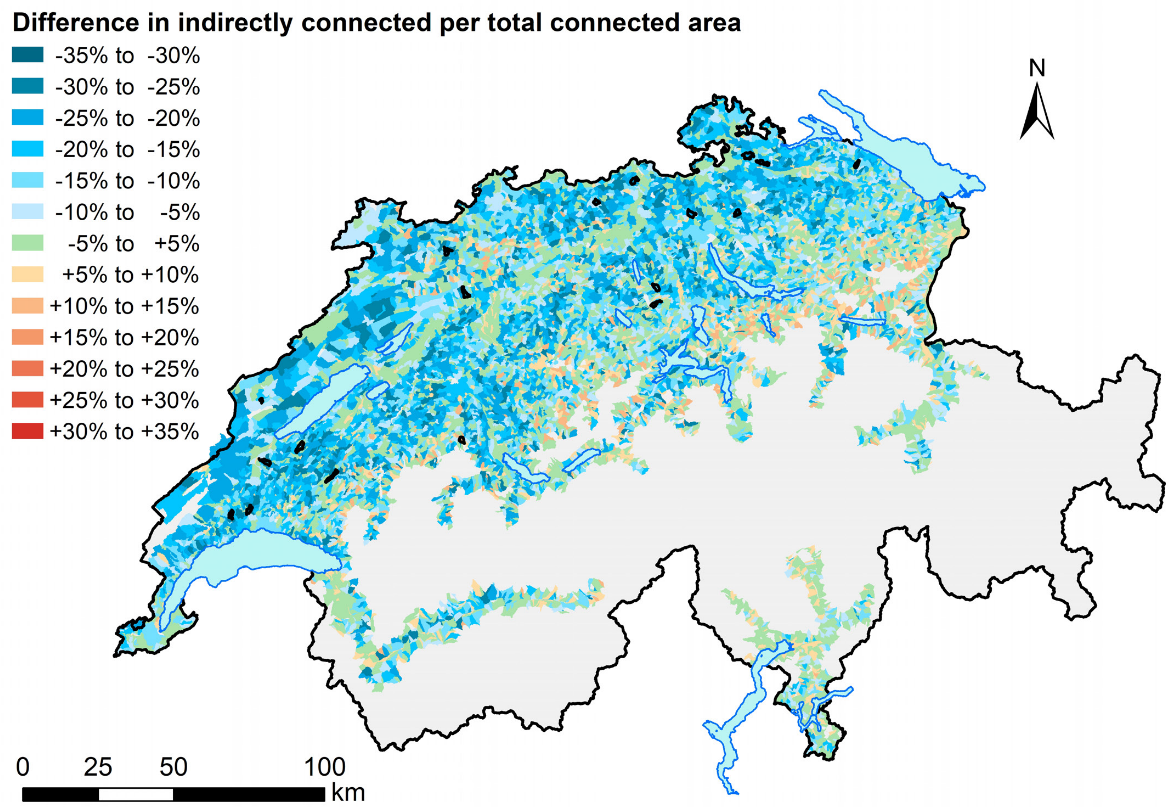


Figure S44: Difference between the fractions of indirectly connected per total connected area reported by the NSCM and the NECM ($f_{NSCM,fracindir}$ - $f_{NECM, fracindir}$). Source of background map: Swisstopo (2010)

S.3. Supporting Information Chapter 3

Are spray drift losses to agricultural roads more important for surface water contamination than direct drift to surface waters?

S3.1 Methods

S3.1.1 Definition of road areas

Road areas were derived from the dataset TLM_STRASSE of the topographic landscape model swissTLM3D (Swisstopo, 2020b). Since this dataset only provides line data for roads, a buffer around road lines was added to generate a dataset of road polygons. The buffer width around each road segment was chosen based on the road categories as defined by the swissTLM3D. The range of road widths covered by each category, and the buffer widths used in this study are shown in Table S10. Afterwards, the resulting road polygon dataset was complemented with additional sealed traffic areas (parking lots and motor way stations) from the polygon dataset TLM_VERKEHRSBAUTE_PLY of the swissTLM3D model.

Table S10: Road categories and buffer widths used for creating a polygon dataset from the road line dataset.

Category	Range of widths according to swissTLM3D	Buffer width used (estimated width / 2)
2 m road	1.81 - 2.80 m	2.3 m / 2 = 1.15 m
3 m road	2.81 - 4.20 m	3.5 m / 2 = 1.75 m
4 m road	4.21 - 6.20 m	5.2 m / 2 = 2.6 m
6 m road	6.21 - 8.20 m	7.2 m / 2 = 3.6 m
8 m road	8.21 m - 10.20 m	9.2 m / 2 = 4.6 m
10 m road	> 10.20 m	10.2 m / 2 = 5.1 m
Highways, motorways	not defined	10.2 m / 2 = 5.1 m
Other roads	< 1.80 m	0 m

S3.1.2 Definition of surface water areas

To determine the surface water areas in our study sites, we combined two datasets of the swissTLM3D (Swisstopo, 2020b) model. Dataset F represents streams (TLM_FLIESSGEWAESSER, line dataset), from dataset B (TLM_BODENBEDECKUNG, polygon dataset) larger surface waters such as large streams, lakes, ponds, and swamps were extracted. Since the stream dataset (F) consists of line data, a buffer around streams was added to generate a dataset of stream polygons. This was only done for smaller streams, since larger streams are covered by the polygons of dataset B. To determine the buffer widths, we measured the width of each stream segment three times using aerial images with a resolution of 0.1 m (Swisstopo, 2019b). The buffer width was then defined as half of the average width measured. For stream segments not visible on the aerial images (e.g. due to coverage by trees), we used the widths determined for the closest downstream segment for which a measurement was available. If no measured downstream segment was available, we used the closest measured upstream segment. If no measured upstream segment was available, we set the buffer width to 1 m. This corresponds to a stream width of 2 m and is expected to be an overestimation in most cases. Finally, the stream polygons resulting from dataset F were combined with the polygons of dataset B into one surface water area dataset.

S3.1.3 Spray drift model: Additional example

To improve the understandibility of the spray drift model described in Sect. 3.2.2.3, we provide an additional example for the calculation of the distance along the wind line $d_{FHT,i,p,w}$ (m). This distance is used for the calculation of the drift reduction factor of forest, hedges and trees $f_{FHT,i,p,w}$ (see eq. 3.4). The additional example (Figure S45), aims on illustrating how the distance along the wind line $d_{FHT,i,p,w}$ is calculated if multiple polygons of forest, hedges and trees (FHT) are located between the non-target area and the sprayed plot.

Between the plot p = 1 and the non-target area cell i = 17, only one FHT polygon is located. Therefore, the FHT distance along the wind line equals the distance along the wind line of this single polygon:

$$d_{FHT,17,1,w} = d_{FHT,A} \tag{S3.1}$$

Between the plot p = 2 and the non-target area cell i = 17, two FHT polygons are located. Therefore, the FHT distance along the wind line is calculated as the sum of these two polygons:

$$d_{FHT,17,2,w} = d_{FHT,A} + d_{FHT,B} \tag{S3.2}$$

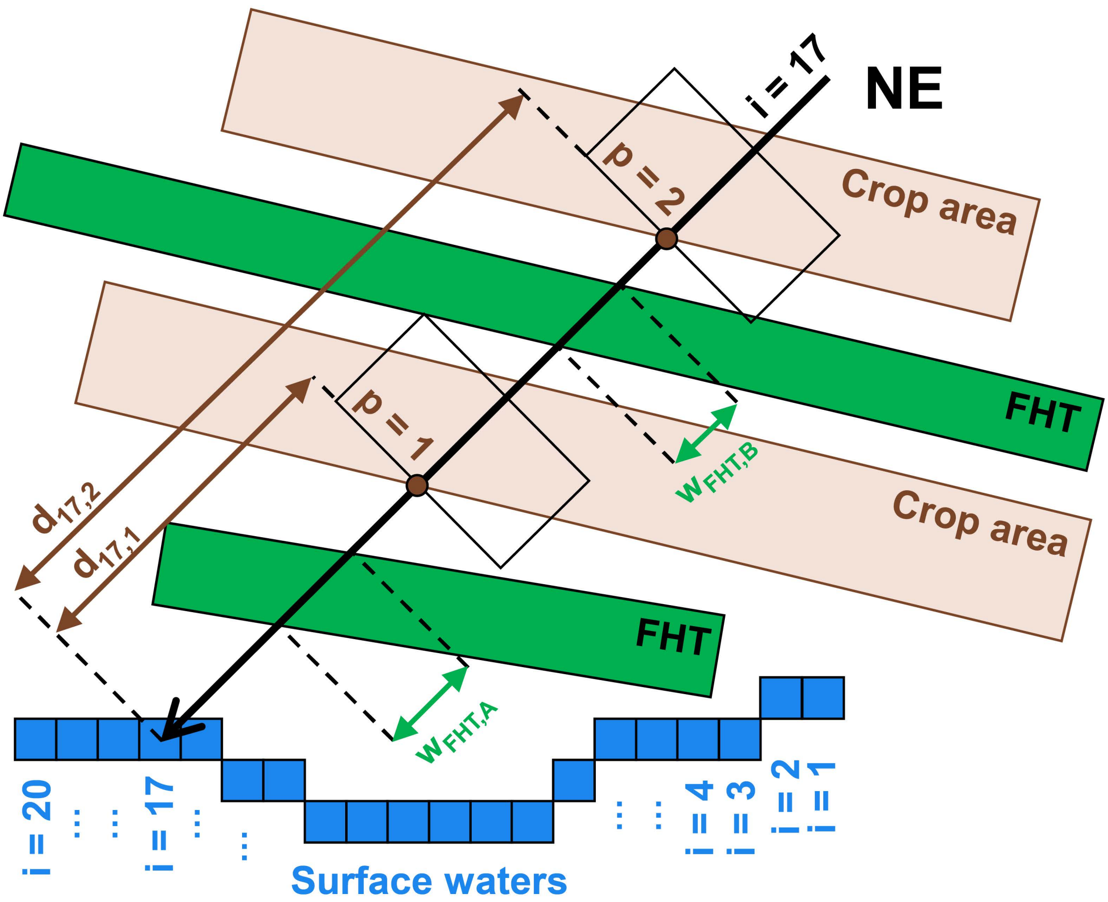


Figure S45: Example of the calculation of drift distances $d_{i,p}$ and barrier distances $d_{FHT,i,p}$ for the non-target area cell i = 17 for the wind direction northeast (NE). In this example, two different polygons of forest, hedges and trees (FHT) act as a barrier.

S3.2 Results

The results of the local model sensitivity analysis are shown in Figure S46 for arable land sites and in Figure S47 for vineyard sites.

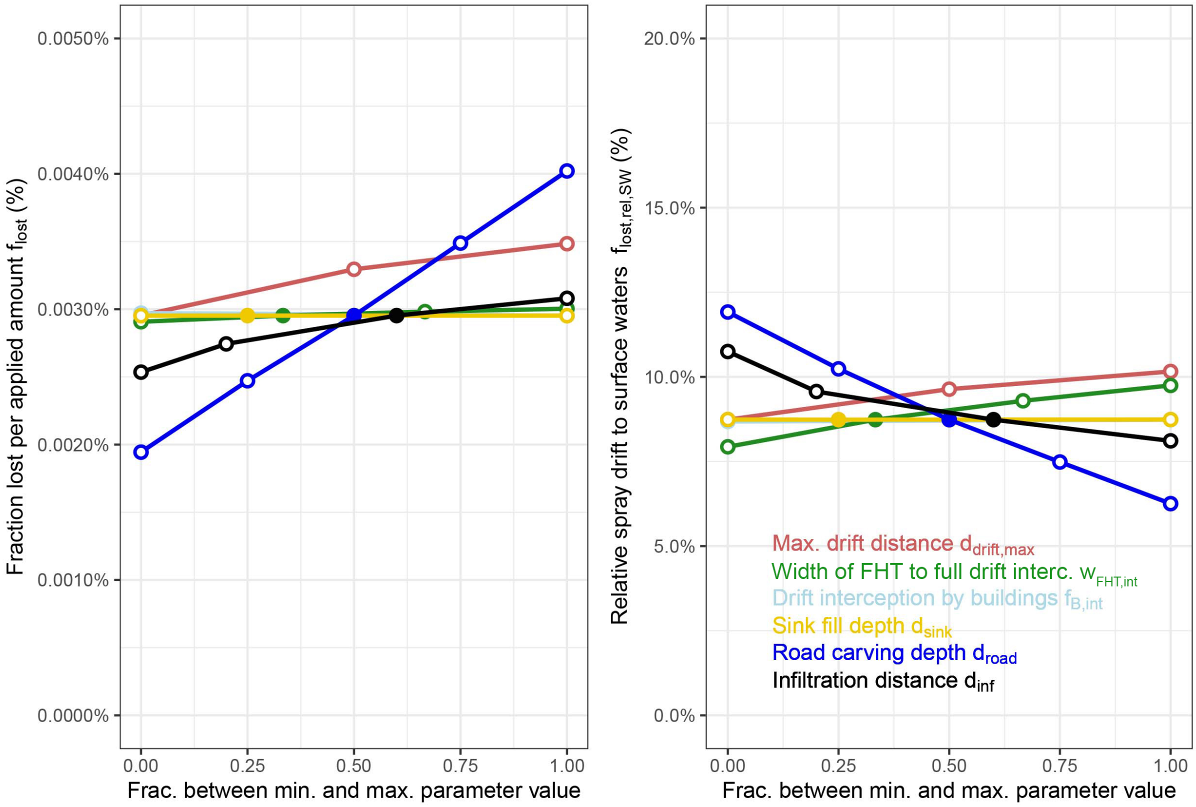


Figure S46: Model sensitivity on parameter changes for arable land sites. Left: Sensitivity of fraction lost per applied amount (f_{lost}). Right: Sensitivity of relative spray drift deposited on surface waters ($f_{lost,rel,SW}$). Filled dots represent the results of the reference parameter set.

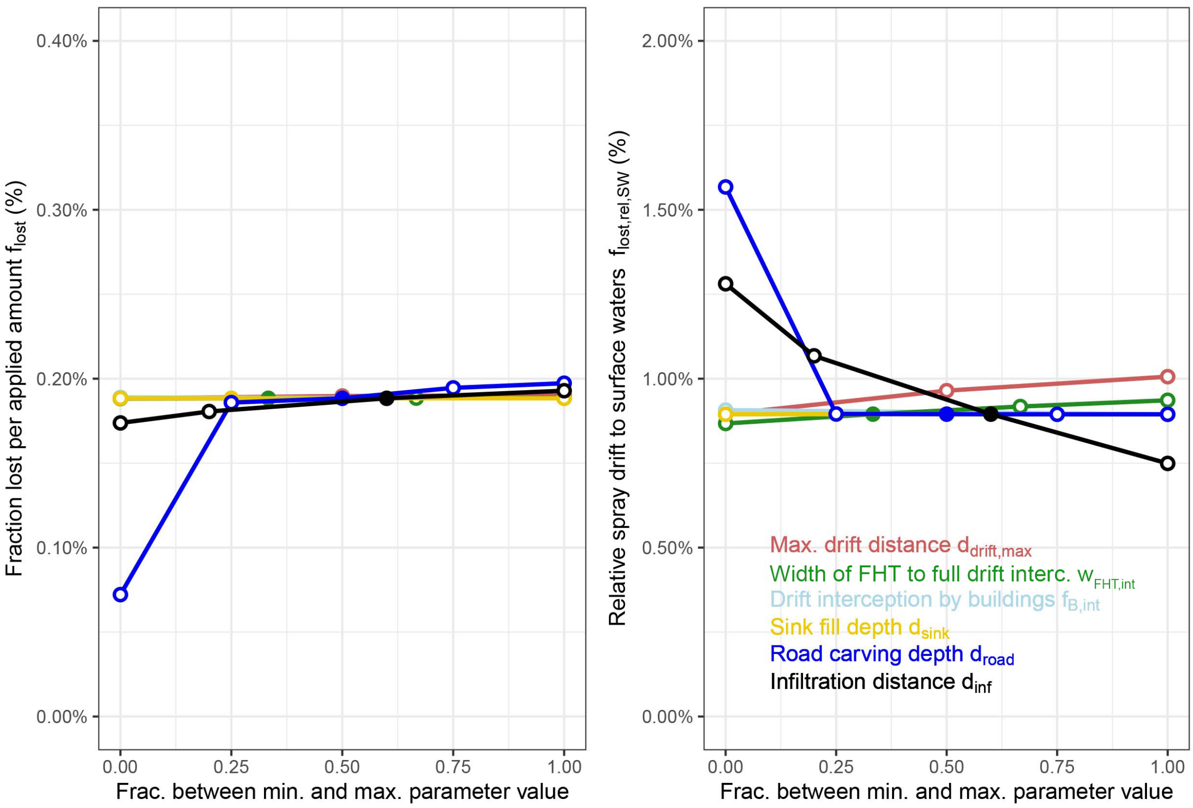


Figure S47: Model sensitivity on parameter changes for vineyard sites. Left: Sensitivity of fraction lost per applied amount (f_{lost}). Right: Sensitivity of relative spray drift deposited on surface waters ($f_{lost,rel,SW}$). Filled dots represent the results of the reference parameter set.

S.4. Supporting Information Chapter 4

Pesticide concentrations in agricultural storm drainage inlets of a small Swiss catchment

The datasets generated and analysed in this study (e.g. pesticide concentrations, rainfall data, discharge data) can be downloaded from the Eawag Research Data Institutional Repository.

DOI: 10.25678/0005X4

S4.1 Methods

S4.1.1 Field work

S4.1.1.1 Sampling sites

Figure S48: Sampling site I1. The inlet is situated between a rather flat farm track and a wheat field with moderate slope. The field is separated by a buffer strip of approximately 6 m width from the farm track.

Figure S49: Sampling site I2. The inlet is situated between a flat farm track and a flat sugar beet field. The field is separated from the farm track with a buffer strip of approximately 0.5 m width. The inlet itself is located on the buffer strip and therefore lies directly at the border of the field and the farm track.

Figure S50: Sampling site I3. The site is situated between a steep einkorn wheat field and a steep farm track. The inlet is separated from the farm track by a grass buffer of approximately 0.5 m width. The field is separated from the farm track by a buffer strip of approximately 2 m width.

Figure S51: Sampling site I4. The site is located at a flat farm track below a steep corn field (left), and next to a flat potato field (right). The two fields are separated from the farm track by a grass buffer strip of approximately 1 m.

Figure S52: Picture of an inlet in the catchment. For taking the picture, the gridded lid was removed. The outlet pipe visible is the only pipe in the inlet, and drains to the stream. The water in the inlet stagnates at the height of the outlet pipe bottom.

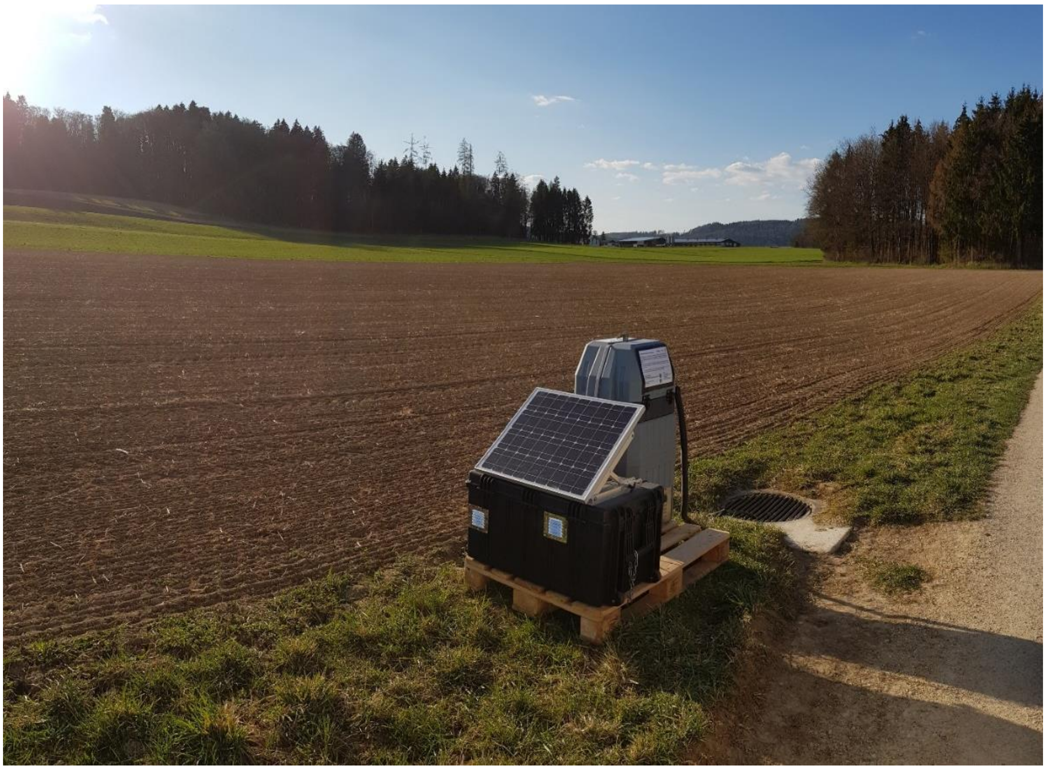


Figure S53: Outside view of sampling site CS.

Figure S54: Inside view of sampling site CS.

Figure S55: Sampling site ST.

S4.1.1.2 Water-level proportional samplers

In the following, we provide a short description of the water-level proportional samplers used in the storm drainage inlets. A detailed description of the samplers is provided in Schönenberger et al. (2020). The water-level proportional samplers consisted of a glass bottle with a volume of 1 L (DURAN Weithalsglasflasche GLS 80), sealed with a screw cap (DURAN GLS80) which had two openings (Figure S). One of the openings was equipped with a bent metal tube, the other one with a plastic tubing of 2 m length (FESTO PUN 6x1-BL) connected to a needle valve (Bronkhorst precision valve, NV-004-HR).

During rain events, surface runoff entering the inlets produces a rise of the water level in the inlets. When the water level was high enough such that the samplers are submerged (this was the case at a water level of 2 cm for inlets with little runoff, and 3 cm for inlets with larger runoff), water starts to flow into the glass bottle (A) through the metal tube (C). In the bottle, the air is compressed and pressed out of the bottle through the needle valve (E). Consequently, an equilibrium between the inflowing water volume, the outflowing air volume, and the compression of air and water in the bottle is established. As soon as this equilibrium is established, an increase of water level pressure leads to an increase in the sampling rate, and consequently, the sampling rate is proportional to the water level. The sampling stops either when the water level drops below the water inlet, or when the sampling bottle is full.

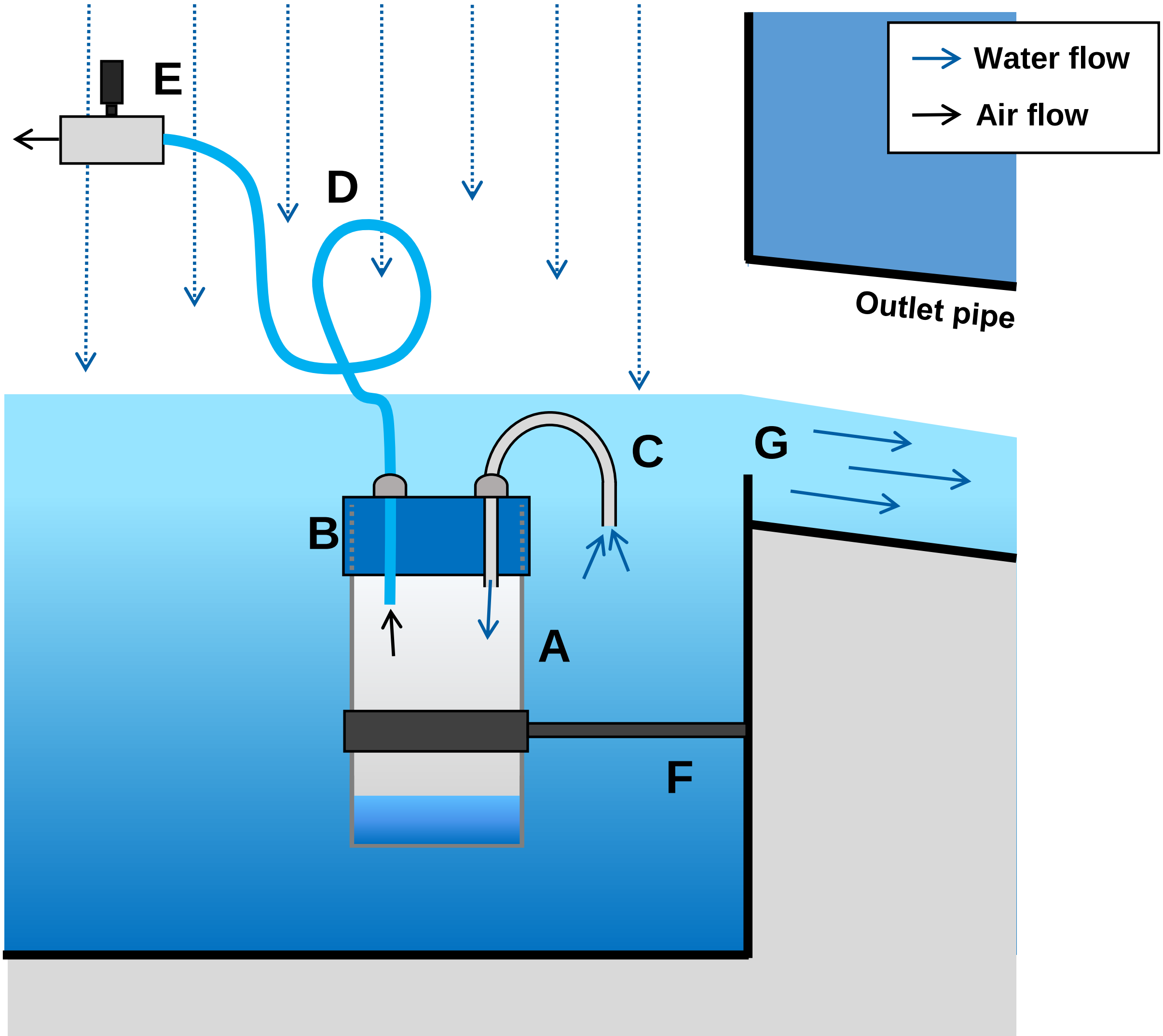


Figure S56: Water-level proportional sampler in a stormwater drainage inlet during a rain event. A: Glass bottle, B: Screw cap, C: Metal tube, D: Plastic tubing, E: Needle valve. F: Fixation of the sampler. G: Weir. Adapted from Schönenberger et al. (2020).

S4.1.1.3 Sampling strategy

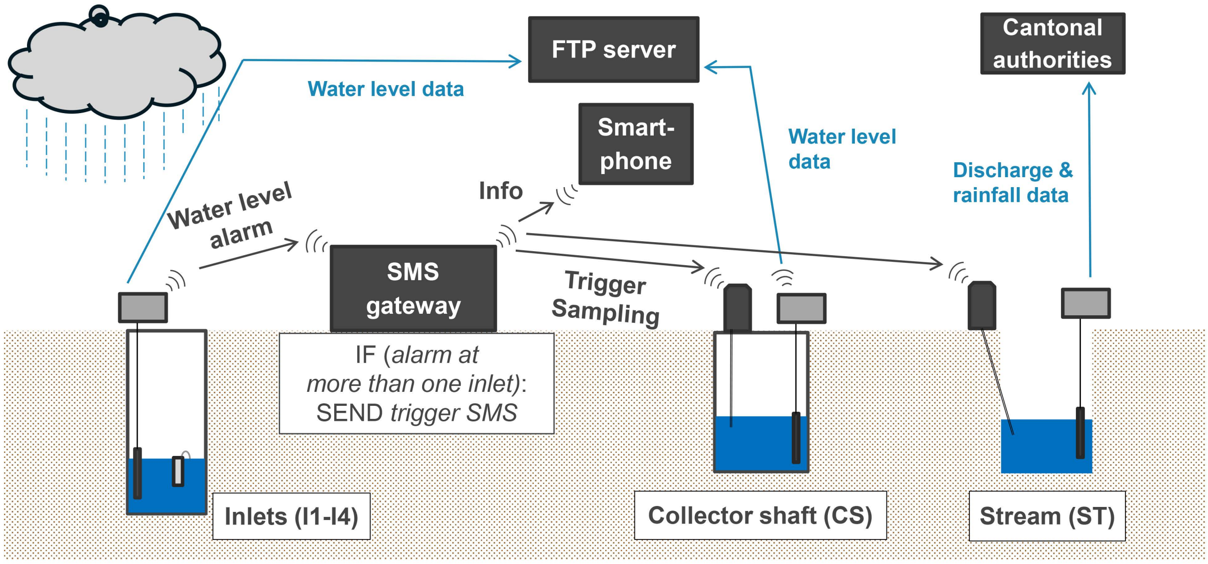


Figure S57: Illustration of the event-based sampling strategy. When the water level threshold was exceeded in at least two of the inlets (I1-I4), the automatic samplers at the collector shaft (CS) and the stream (ST) were triggered via the GPRS module to start sampling. Additionally, water level data and the information about the triggering of the samplers were sent to the research institute via the GPRS modules.

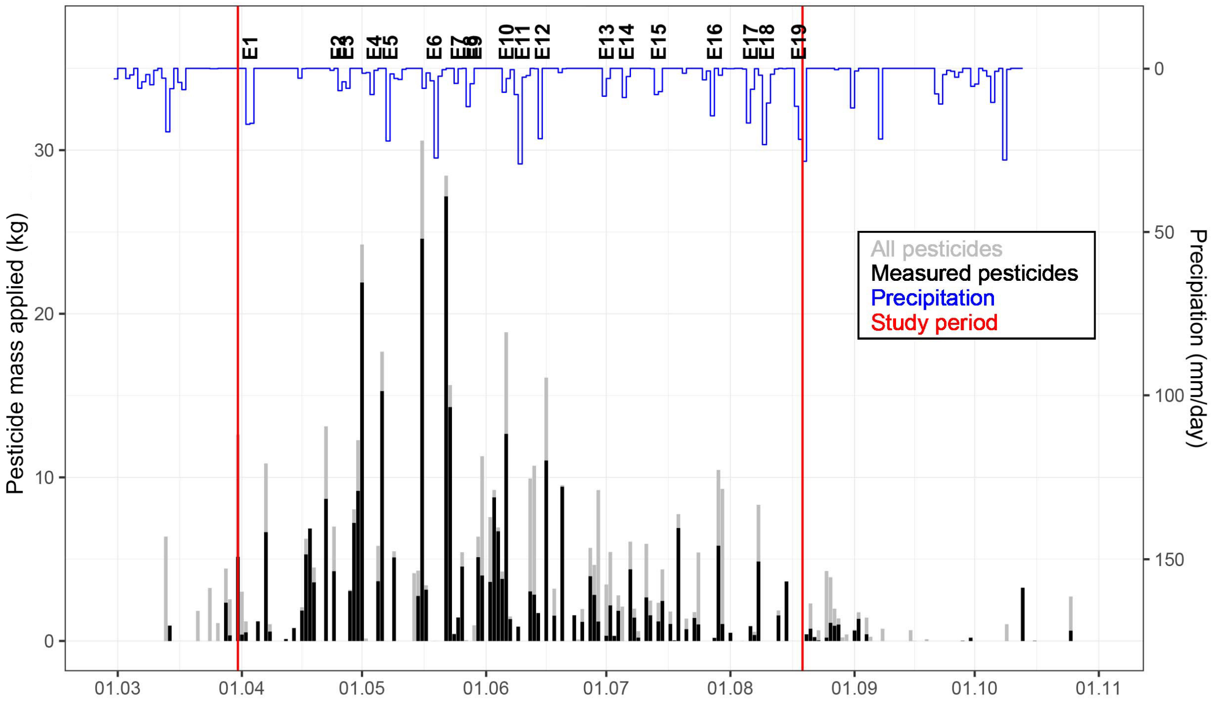


Figure S58: Total masses of pesticides applied in the study catchment per day in 2019 (kg). The red lines depict the start and the end of the study period (01.04.2019 and 20.08.2019). Grey bars show the total pesticide mass applied on the respective day. Black bars show the total pesticide mass applied for only those substances that were analysed within this study. Oils used as pesticides (e.g. paraffin oil, rapeseed oil) were excluded from the analysis. E1 to E19 indicate the rain events sampled in this study.

S4.1.1.4 Field mapping

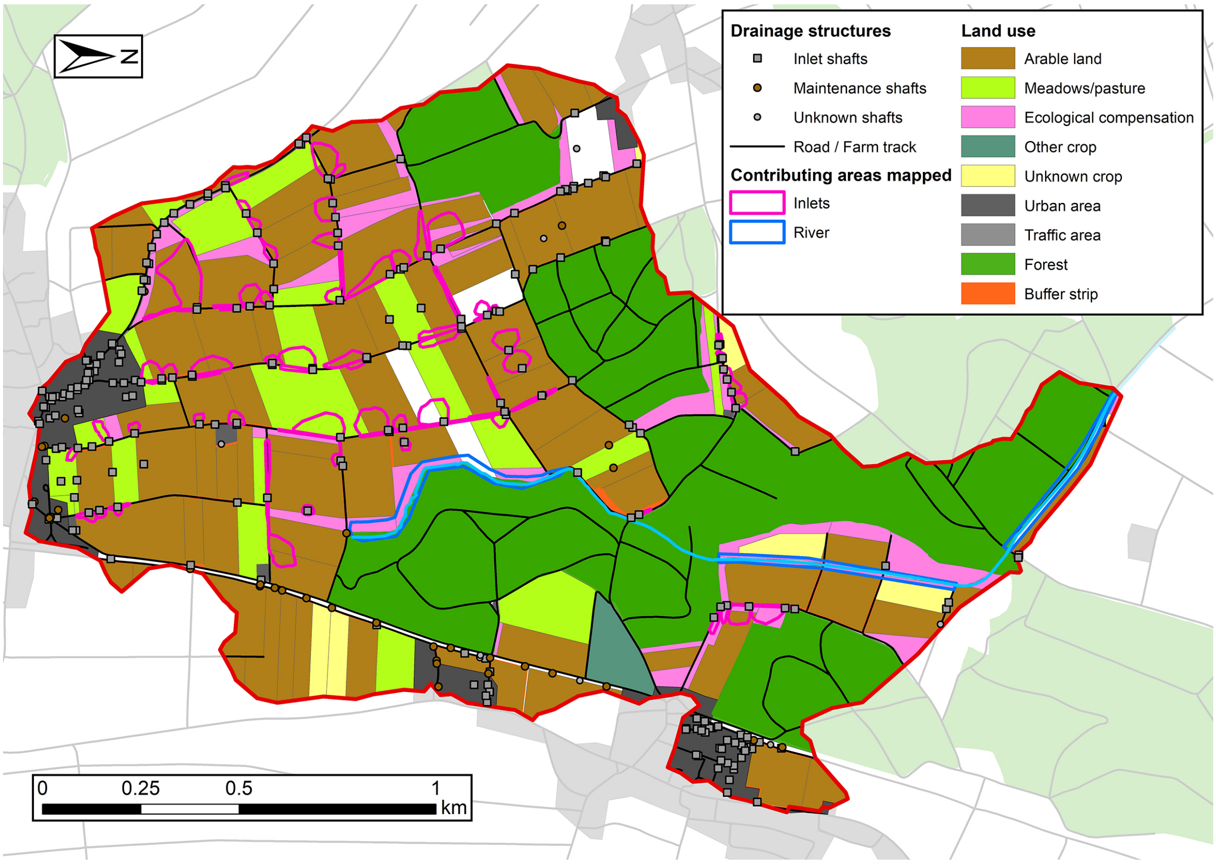


Figure S59: Map of surface runoff flow paths during a snowmelt event on 12 March 2018. The mapped contributing areas are minimal contributing areas and may be much larger in reality. They were only mapped for some of the inlets and may differ for other events. Source of background map: Swisstopo (2020b).

S4.1.2 Chemical analysis

In the following, further details on the chemical analysis procedure are given. A table with all substances measured is given below (Table S13).

Field samples were stored at -20° until further process. After thawing, the sample was shaken, and 1.5 ml sample was transferred to a 1.5 ml vial and closed with a magnetic starburst cap. The sample preparation was achieved through a fully automated workflow using a PAL RTC (CTC analytics AG, Switzerland) equipped with a dilutor tool, centrifuge, C-stack, fast wash station and an injection valve. After centrifugation of the samples (5 min at 2000g), 600 µL of supernatant was aspirated by the dilutor. The dilutor needle was washed by aspirating 10 µL of nanopure water at the fast wash station. Afterwards, 10 µL of a standard mix containing 84 isotopic labelled internal standards (ISTD, details Table S13 at a concentration of 0.01 mg/L was added to the dilutor tubing and separated again with 10 µL of nanopure water. Depending on the sample type either an exact volume of standard solution (concentration: 0.06, 0.006 and 0.0006 mg/L) was added and then equalized by an ethanol volume ranging from 0 to 50 µL (standards and spiked samples) or just the equalization volume of 50 µL Ethanol was added (samples and blanks). This ensured constant sample constitution and an organic content of ~5%. The entire sample volume of 670 µL was then transferred into an empty vial equipped with a slitted septa and mixed by aspirating and dispensing the dilutor. Sample preparation occurred interlaced with sample acquisition. During one measurement four samples were prepared as described above.

For the measurement, a volume of 100 µL was injected on to the chromatographic system. Chromatographic separation was achieved using a reversed phase column (Atlantis T3, particle size 3µm, 3.0×150 mm, Waters) and a linear water-methanol gradient, both acidified with 0.1% formic acid. The flow rate was 0.3 µL/min and the column temperature was 30°C. The gradient was as follows: 0-1.5 min constant at 0% methanol, 1.5-18.5 min linear gradient to 95% methanol, 18.5-30.5 min constant at 95% methanol followed by equilibration (0% methanol) for 3.5 min. Data acquisition was accomplished with a Lumos Fusion (Thermo Fisher Scientific) running in positive and negative electrospray ionization mode separately (spray voltage: 3500 V in pos, 3000 V in neg). Full scans were recorded with a resolution of 240'000 (at m/z 200) and mass range 100-1000 m/z followed by three data-dependent MS2 scans using higher energy collision-induced dissociation (HCD) at a resolution of 15'000 (at m/z 200).

Peak integration was performed using TraceFinder 5.1 with a mass tolerance of 5 ppm. Substance confirmation occurred through comparison of the retention time, exact mass and fragment spectra with reference material. Quantification was achieved with a linear calibration curve using the peak area ratio of the analyte and ISTD. The calibration curve ranged from 10 to 5000 ng/L. For compounds without structurally identical ISTD, a closely eluting or structurally similar ISTD was chosen to reach the best relative recovery (close to 100% in spiked samples). The assignment of ISTDs and relative recoveries are shown in Table S13. For those compounds, the concentrations were corrected by the relative recovery. The LOQ was determined by the concentration of the lowest standard that was still detected with a good chromatographic peak (at least 4 sticks) and whose area was at least 4 times higher than in laboratory or field blank samples. The lowest calibrations standard value was then corrected by the matrix factor for the final LOQ. For 80% of the compounds, the LOQ was 20 ng/L or lower (see Table S13). For quality control, 54 laboratory and 11 field blanks were measured and taken into account for the LOQ. Additionally, 18 random samples were spiked with 50 and 500 ng/L to determine the relative recovery and matrix suppression.

S4.1.3 Data analysis

S4.1.3.1 Surface runoff connectivity model

Table S11: Parameters of the surface runoff connectivity model used for determining catchments of inlets, river, and internal sinks.

Parameter	Value
Hedge infiltration	No hedges in the catchment
Forest infiltration width	No infiltration in forests
Road carving depth	25 cm
Sink depth	25 cm
Shortcut definition	Only inlets act as shortcut
Maximal flow distance	No restriction on maximal flow distance

S4.1.3.2 Discharge measurement in inlets

As mentioned in Sect. 4.2.4.4, the discharge in the inlets was determined using water level measurements combined with triangular weirs for which a rating curve was calibrated. The weirs consisted of stainless chromium steel plates with two triangles of different slopes cut out (see Figure S60), and were installed in front of the outlet pipes of the measured inlets. The space between the outlet pipe and the weir was sealed with rubber.

For determining the rating curve of the weirs, their wetted area was split into three areas A, B, and C, as shown in Figure S60. For each area, a separate rating curve was determined and the rating curve of the weir was calculated by summing up the contributions of all three areas (eq. S4.4 to S4.6).

Area A was defined as the wetted area for water levels (p) smaller or equal to the water level at the slope changing point of the triangular weir (p_{wc}). For this area, the weir corresponds to a normal triangular weir and its rating curve can be described according to eq. S4.1 (Aigner 2008). Area B was defined as the wetted area between the slope changing point of the triangular weir (p_{wc}) and the water level up to which the discharge was calibrated ($p_{cal,max}$). We neglected the influence of flow in area A on the flow in area B and assumed that the shape of the rating curve of area B corresponded to the curve of a trapezoid weir (eq. S4.2, Aigner 2008).

For area C (water levels higher than $p_{cal,max}$), we created three different assumptions, corresponding to a minimum (Q_{min}), moderate (Q_{mod}) and high (Q_{high}) discharge estimate (example, see Figure S61). For the minimum estimate (eq. S4.4), we set the upper discharge limit to the maximal discharge for which the weir was calibrated $Q(p_{cal,max})$. For the moderate estimate (eq. S4.5), we assumed the shape of the rating curve in area C to correspond to the curve of a circular weir (eq. S4.3, Aigner 2008). The upper discharge limit was set to the discharge calculated for the water level at the upper weir end (p_{max}). For the high estimate (eq. S4.6), we extrapolated the shape of the rating curve of area B (eq. S4.2) and set an upper discharge limit to the discharge calculated for p_{max}.

The weir discharge coefficients μ_{tri} and μ_{tra} were calibrated by pouring known discharges into an inlet with a tube and measuring the emerging water levels. Since no discharges corresponding to water levels higher than $p_{cal,max}$ could be produced with the tube, the coefficient μ_{cir} was calibrated using only one data point (i.e. the data point at water level $p_{cal,max}$).

$$Q_{tri}(p) = \frac{8}{15}\mu_{tri} \cdot \sqrt{2g} \cdot \frac{b}{2 \cdot p_{wc}} \cdot p^{\frac{5}{2}} \tag{S4.1}$$

$$Q_{tra}(p) = \frac{2}{3}\mu_{tra} \cdot \sqrt{2g} \cdot b \cdot (p - p_{wc})^{\frac{1}{5}} \cdot \left(1 + \frac{4}{5}\frac{(p-p_{wc}) \cdot w}{b \cdot h}\right) \tag{S4.2}$$

$$Q_{cir}(p) = \mu_{cir} \cdot \sqrt{2g} \cdot d^{\frac{2}{3}} \cdot \left(\left(p - p_{cal,max} + r\right)^{\frac{11}{6}} - r^{\frac{11}{6}}\right) \tag{S4.3}$$

$$Q_{min} = \begin{cases} Q_{tri}(p) & | \; p \leq p_{wc} \\ Q_{tri}(p_{wc}) + Q_{tra}(p) & | \; p_{wc} < p \leq p_{cal.max} \\ Q_{tri}(p_{wc}) + Q_{tra}(p_{cal.max}) & | \; p > p_{cal.max} \end{cases} \tag{S4.4}$$

$$Q_{mod} = \begin{cases} Q_{tri}(p) & | \; p \leq p_{wc} \\ Q_{tri}(p_{wc}) + Q_{tra}(p) & | \; p_{wc} < p \leq p_{cal.max} \\ Q_{tri}(p_{wc}) + Q_{tra}(p_{cal.max}) + Q_{cir}(\min(p, p_{max})) & | \; p > p_{cal.max} \end{cases} \tag{S4.5}$$

$$Q_{high} = \begin{cases} Q_{tri}(p) & | \; p \leq p_{wc} \\ Q_{tri}(p_{wc}) + Q_{tra}(p) & | \; p_{wc} < p \leq p_{cal.max} \\ Q_{tri}(p_{wc}) + Q_{tra}(\min(p, p_{max})) & | \; p > p_{cal.max} \end{cases} \tag{S4.6}$$

with:

Q_{tri}, Q_{tra}, Q_{cir}:	Rating curves for the triangular, trapezoid, and circular part of the weir (m^3 s^{-1})
Q_{min}, Q_{mod}, Q_{high}:	Minimal, moderate, and high discharge estimate (m^3 s^{-1})
p:	Water level (m)
p_{wc}:	Water level at the slope changing point of the triangular weir (= 0.03 m)
$p_{cal,max}$:	Maximal water level up to which the weir was calibrated (m)
p_{max}:	Water level at the upper end of the weir (= 0.075 m)
b, h, w:	Dimensions of the triangular weir (m) (see Figure S)
d:	Diameter of the outlet pipe (m)
r:	Radius of the outlet pipe (m)
g:	Acceleration due to gravity (= 9.807 m s^{-2})
μ_{tri}, μ_{tra}, μ_{cir}:	Weir discharge coefficients (-)

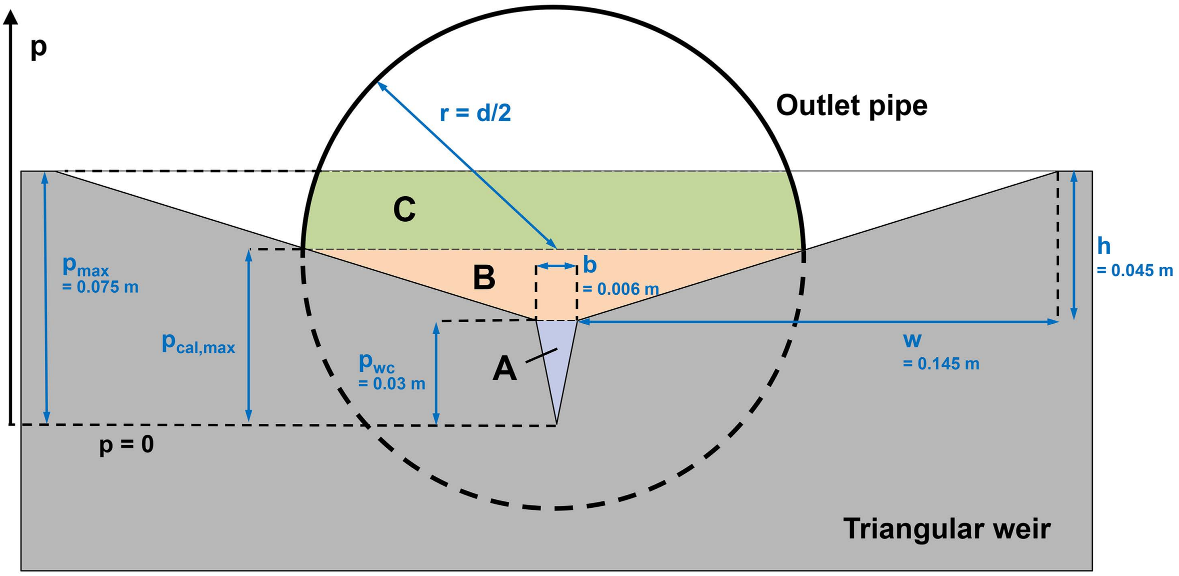


Figure S60: Dimensions of the triangular weir (grey area) and the subareas used for rating curve determination.

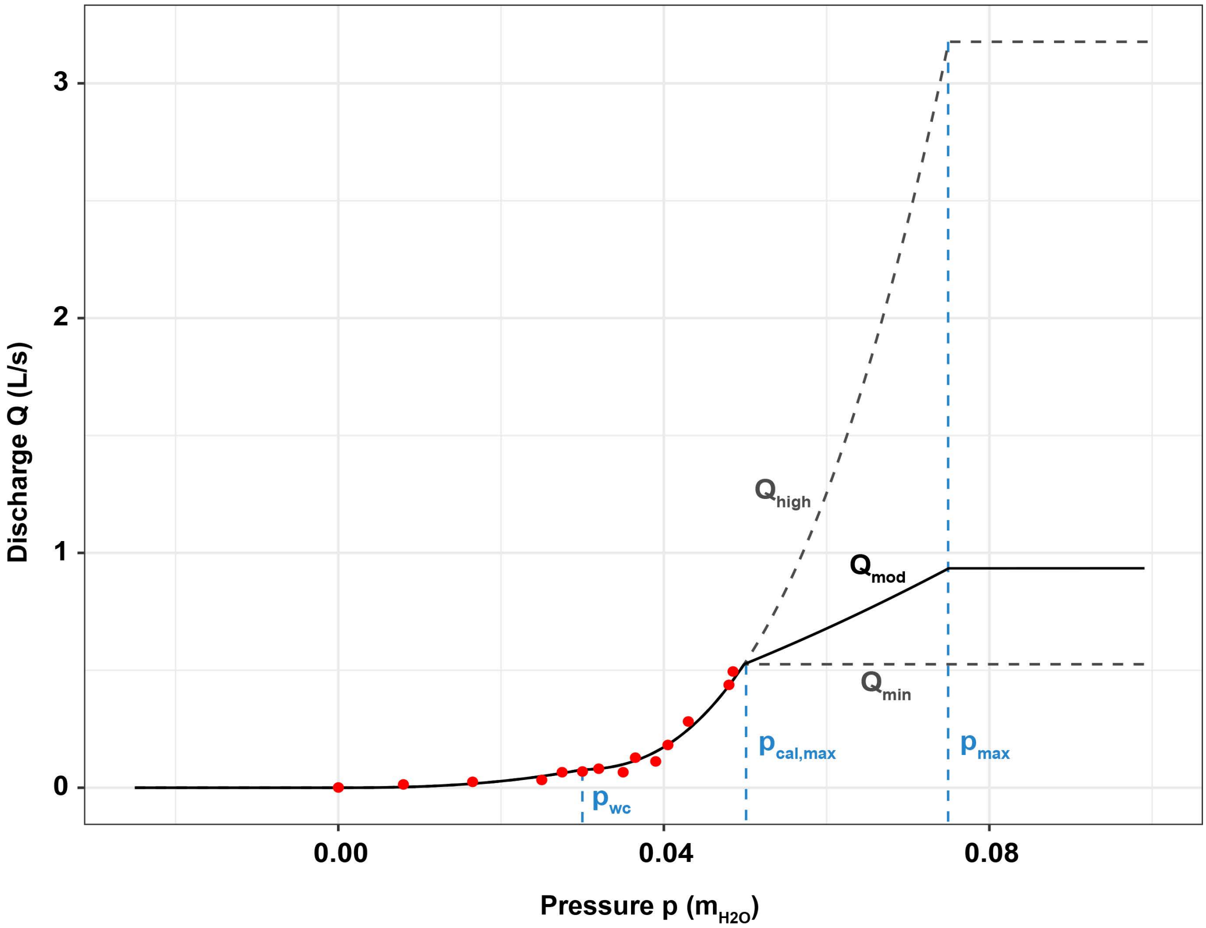


Figure S61: Rating curve of the triangular weir installed in inlet 3. The black solid line represents the moderate discharge estimate (Q_{mod}) and the black dashed lines represent the minimal and high discharge estimates (Q_{min} and Q_{high}). The red dots show the measurements used for calibration of the rating curve.

To compare the discharge in the inlets and the stream, we calculated the ratio between the discharge sum of all four inlets to the discharge in the stream (r_Q) (eq. S4.7). For the discharge measured in the stream Q_{stream}, the cantonal authorities provided no information on uncertainty. Expecting that the relative uncertainty of the discharge through inlets is much larger than the uncertainty in stream discharge, we neglected the latter.

$$\mathbf{r_Q} = \begin{pmatrix} r_{Q,min} \\ r_{Q,mod} \\ r_{Q,high} \end{pmatrix} = \frac{\sum_{i=1}^{4} \mathbf{Q_{inl,i}}}{Q_{stream}} = \frac{\sum_{i=1}^{4} \begin{pmatrix} Q_{inl,i,min} \\ Q_{inl,i,mod} \\ Q_{inl,i,high} \end{pmatrix}}{Q_{stream}} \qquad \text{(S4.7)}$$

$\mathbf{Q_{inl,i}}$: Discharge estimates (minimum, moderate, high) in inlet i ($m^3\ s^{-1}$)

Q_{stream}: Discharge in the stream ($m^3\ s^{-1}$)

Additionally, we calculated the ratio between the discharge sum in inlets and the fast discharge in the stream ($r_{Q,fast}$) (eq. S4.8).

$$\mathbf{r_{Q,fast}} = \begin{pmatrix} r_{Q,fast,min} \\ r_{Q,fast,mod} \\ r_{Q,fast,high} \end{pmatrix} = \frac{\sum_{i=1}^{4} \mathbf{Q_{inl.i}}}{\mathbf{Q_{stream,fast}}} = \frac{\sum_{i=1}^{4} \begin{pmatrix} Q_{inl,i,min} \\ Q_{inl,i,mod} \\ Q_{inl,i,high} \end{pmatrix}}{\begin{pmatrix} Q_{stream,fast,high} \\ Q_{stream,fast,mod} \\ Q_{stream,fast,low} \end{pmatrix}} \qquad \text{(S4.8)}$$

$\mathbf{Q_{stream,fast}}$: Fast discharge fraction estimates in the stream ($m^3\ s^{-1}$)

We estimated the fast discharge fraction in the stream using a recursive filter technique (Lyne and Hollick, 1979) for discharge separation (function "BaseflowSeparation" of the R package "EcoHydRology", version 0.4.12.1, Fuka et al. (2018)). We used three different filter parameters (0.9, 0.925, and 0.95; see Nathan and Mcmahon (1990)) to come up with a low, moderate, and high estimate of the fast discharge fraction.

Based on the discharge measurements in the four inlets, we estimated the total discharge flowing through all inlets in the catchment $Q_{inl,tot}$. For this, we used three simple extrapolation methods. In the first two methods, we assumed that the discharge in an inlet is proportional to the road area (eq. S4.9) or to the agricultural area connected to the inlet (eq. S4.10). In the third method, we assumed that the discharge is proportional to the number of inlets (eq. S4.11). These three methods are meant to provide a rough estimate of the total discharge and various parameters influencing the total discharge (such as slope, soil permeability, crop types, spatial distribution of rainfall) were not taken into account here.

$$\mathbf{Q_{inl,tot,road}} = \frac{\sum_{i=1}^{4} A_{road,i}}{A_{road,tot}} \cdot \sum_{i=1}^{4} \mathbf{Q_{inl,i}} \qquad \text{(S4.9)}$$

$A_{road,i}$: Road area connected to inlet i (m^2)

$A_{road,tot}$: Total road area connected to inlets in the catchment (m^2)

$Q_{inl,i}$: Discharge in inlet i ($m^3\ s^{-1}$)

$$\mathbf{Q_{inl,tot,agri}} = \frac{\sum_{i=1}^{4} A_{agri,i}}{A_{agri,tot}} \cdot \sum_{i=1}^{4} \mathbf{Q_{inl,i}} \qquad \text{(S4.10)}$$

$A_{agri,i}$: Agricultural area connected to inlet i (m^2)

$A_{agri,tot}$: Total agricultural area connected to inlets in the catchment (m^2)

$$\mathbf{Q_{inl,tot,num}} = \frac{n_{inl,tot}}{n_{inl,measured}} \cdot \sum_{i=1}^{4} \mathbf{Q_{inl,i}} = \frac{158}{4} \cdot \sum_{i=1}^{4} \mathbf{Q_{inl,i}} \qquad \text{(S4.11)}$$

$n_{inl,\ measured}$: Number of inlets with discharge measurements (-)

$n_{inl,\ tot}$: Total number of inlets in the catchment (-)

S4.1.3.3 Model of concentrations in inlets

Table S12: Overview over the variables used for building the linear mixed model.

Variable	Abbre-viation	Type	Discrete/ Continuous	Unit	Range/ Categories
Inlet concentration	$\log_{10}(c)$	Response variable	Continuous	ng L^{-1}	$\log_{10}([5, 62000])$
Time since application	t_{appl}	Fixed effect	Continuous	days	[1.2, 142]
Amount of pesticide applied per area	$\log_{10}(m_{appl})$	Fixed effect	Discrete	g ha^{-1}	$\log_{10}([1.2, 1600])$
Freundlich adsorption coefficient normalized to organic carbon content	$\log_{10}(K_{foc})$	Fixed effect	Discrete	mg L^{-1}	$\log_{10}([20, 4900])$
Octanol-water partition coefficient	$\log_{10}(K_{ow})$	Fixed effect	Discrete	-	[-1.2, 4.7]
Half-life in water	$DT_{50,\ water}$	Fixed effect	Discrete	days	[0.30, 92]
Half-life in soil	$DT_{50,\ soil}$	Fixed effect	Discrete	days	[0.34, 500]
Moderate estimate of the discharge in the inlet during the event	$\log_{10}(Q_{mod})$	Fixed effect	Continuous	L	$\log_{10}([0,8500])$
Potential transport processes involved	$p_{transport}$	Fixed effect	Categorial	-	(A, B, C, D) (see Sect. 4.2.4.3)
Inlet sampled	i	Random effect	Categorial	-	(1, 2, 3, 4)

Table S13: List of the 51 substances analysed. AS: Active substance, TP: Transformation product, LOQ: Limit of quantification, RR: Relative recovery, LMM: Linear mixed model. The samples were measured in three sets. Below, we therefore report the LOQs and RRs for each set. Semiquantiative: pt = partially.

Substance measured	InChIKey	Substance type	Pesticide class	LOQ Set 1 (ng/L)	LOQ Set 2 (ng/L)	LOQ Set 3 (ng/L)	RR set 1 (%)	RR set 2 (%)	RR set 3 (%)	Semiquantitative	Internal standard	Used for LMM
Azoxystrobin	WFDXOXNFNRHQEC-GHRIWEEISA-N	AS	Fungicide	7	9	6	78	82	97	no	Azoxystrobin D4	
Bixafen	LDLMOOXUCMHBMZ-UHFFFAOYSA-N	AS	Fungicide	8	20	5	80	98	105	no	Flufenacet D5	x
Boscalid	WYEMLYFITZORAB-UHFFFAOYSA-N	AS	Fungicide	10	10	4	89	92	93	no	Boscalid D4	
Carfentrazone-ethyl	MLKCGVHIFJBRCD-UHFFFAOYSA-N	AS	Herbicide	10	20	15	50	83	74	no	Epoxiconazole D4	
Chlortoluron	JXCGFZXSOMJFOA-UHFFFAOYSA-N	AS	Herbicide	10	6	6	83	97	101	no	Chlortoluron D6	
Cymoxanil	XERJKGMBORTKEO-VZUCSPMQSA-N	AS	Fungicide	9	9	9	76	87	174	pt	Metamitron D5	x
Cyproconazole	UFNOUKDBUJZYDE-UHFFFAOYSA-N	AS	Fungicide	8	8	4	83	100	102	no	Dimethenamid D3	
Difenoconazole	BQYJATMQXGBDHF-UHFFFAOYSA-N	AS	Fungicide	25	8	8	121	106	114	no	Pyraclostrobin D3	x
Diflufenican	WYEHFWKAOXOVJD-UHFFFAOYSA-N	AS	Herbicide	500	200	100	81	143	103	no	Metrafenone D9	
Dimethachlor	SCCDDNKJYDZXMM-UHFFFAOYSA-N	AS	Herbicide	6	9	10	79	86	106	pt	Dimethenamid D3	
Dimethenamid	JLYFCTQDENRSOL-UHFFFAOYSA-N	AS	Herbicide	9	8	4	89	102	98	no	Dimethenamid D3	x
Epoxiconazole	ZMYFCFLJBGAQRS-UHFFFAOYSA-N	AS	Fungicide	7	8	6	75	78	99	no	Epoxiconazol D4	x
Ethofumesate	IRCMYGHHKLLGHV-UHFFFAOYSA-N	AS	Herbicide	9	20	10	88	109	87	no	Azoxystrobin D4	x
Fenpropimorph	RYAUSSKQMZRMAI-ALOPSCKCSA-N	AS	Fungicide	6	5	10	64	105	93	no	Metribuzin D3	x
Florasulam	QZXATCCPQKOEIH-UHFFFAOYSA-N	AS	Herbicide	100	50	20	73	91	95	no	2,4-D3	x
Fluazifop (free acid)	YUVKUEAFAVKILW-UHFFFAOYSA-N	AS	Herbicide	40	9	10	100	103	98	no	Mecoprop D6	
Fluazinam	UZCGKGPEKUCDTF-UHFFFAOYSA-N	AS	Fungicide	200	8	4	345	123	121	no	Fipronil 13C15N2	
Flufenacet	IANUJLZYFUDJIH-UHFFFAOYSA-N	AS	Herbicide	8	20	6	84	98	91	no	Flufenacet D4	
Fluopicolide	GBOYJIHYACSLGN-UHFFFAOYSA-N	AS	Fungicide	8	9	2	97	99	109	no	Dimethenamid D3	
Flupyrsulfuron-methyl	DTVOKYWXACGVGO-UHFFFAOYSA-N	AS	Herbicide	10	9	10	66	105	102	no	Boscalid D4	
Foramsulfuron	PXDNXJSDGQBLKS-UHFFFAOYSA-N	AS	Herbicide	9	9	4	80	93	97	pt	Metribuzin D3	
Iodosulfuron-methyl	VWGAYSCWLXQJBQ-UHFFFAOYSA-N	AS	Herbicide	10	15	10	47	102	100	no	Azoxystrobin D4	x
Isoproturon	PUIYMUZLKQOUOZ-UHFFFAOYSA-N	AS	Herbicide	2	4	6	82	94	103	pt	Isoproturon D5	
Lenacil	ZTMKADLOSYKWCA-UHFFFAOYSA-N	AS	Herbicide	10	15	8	88	100	100	no	Lenacil(cyclohexyl) D4	x
Mandipropamid	KWLVWJPJKJMCSH-UHFFFAOYSA-N	AS	Fungicide	8	9	4	92	108	117	no	Dimethenamid D3	
Mecoprop	WNTGYJSOUMFZEP-UHFFFAOYSA-N	AS	Herbicide	100	40	5	84	109	95	no	Mecoprop D6	x
Mesosulfuron-methyl	RSMUVYRMZCOLBH-UHFFFAOYSA-N	AS	Herbicide	8	8	9	89	99	120	no	Dimethenamid D3	x

Substance measured	InChIKey	Substance type	Pesticide class	LOQ Set 1 (ng/L)	LOQ Set 2 (ng/L)	LOQ Set 3 (ng/L)	RR set 1 (%)	RR set 2 (%)	RR set 3 (%)	Semiquantitative	Internal standard	Used for LMM
Metamitron	VHCNQEUWZYOAEV-UHFFFAOYSA-N	AS	Herbicide	20	7	8	89	105	109	no	Metamitron D5	x
Metolachlor	WVQBLGZPHOPPFO-UHFFFAOYSA-N	AS	Herbicide	8	8	4	91	85	95	no	Metolachlor D6	
Metrafenone	AMSPWOYQQAWRRM-UHFFFAOYSA-N	AS	Fungicide	15	9	6	83	95	93	no	Metrafenone D9	x
Metribuzin	FOXFZRUHNHCZPX-UHFFFAOYSA-N	AS	Herbicide	9	7	4	84	95	99	no	Metribuzin(S-methyl-D3)	x
Napropamide	WXZVAROIGSFCFJ-UHFFFAOYSA-N	AS	Herbicide	8	10	6	71	88	103	no	Terbutylazin D5	
Nicosulfuron	RTCOGUMHFFWOJV-UHFFFAOYSA-N	AS	Herbicide	10	8	6	92	98	92	no	Nicosulfuron D6	
Pencycuron	OGYFATSSENRIKG-UHFFFAOYSA-N	AS	Seed treatment	15	9	6	96	91	105	no	Metolachlor D6	x
Propamocarb	WZZLDXDUQPOXNW-UHFFFAOYSA-N	AS	Fungicide	5	25	50	139	135	110	no	Metribuzin(S-methyl-D3)	
Propiconazole	STJLVHWMYQXCPB-UHFFFAOYSA-N	AS	Fungicide	15	8	6	104	106	106	no	Metrafenone D9	x
Prosulfocarb	NQLVQOSNDJXLKG-UHFFFAOYSA-N	AS	Herbicide	15	8	2	104	86	88	no	Epoxiconazol D4	x
Prothioconazole	MNHVNIJQQRJYDH-UHFFFAOYSA-N	AS	Fungicide	100	200	100	50	74	81	no	Pyraclostrobin D3	
Pyraclostrobin	HZRSNVGNWUDEFX-UHFFFAOYSA-N	AS	Fungicide	50	9	15	81	101	96	no	Pyraclostrobin D3	
Spiroxamine	PUYXTUJWRLOUCW-UHFFFAOYSA-N	AS	Fungicide	100	25	20	153	144	61	no	Nicosulfuron D6	
Tembotrione	IUQAXCIUEPFPSF-UHFFFAOYSA-N	AS	Herbicide	50	8	8	62	111	109	no	Azoxystrobin D4	
Terbuthylazine	FZXISNSWEXTPMF-UHFFFAOYSA-N	AS	Herbicide	9	7	6	85	100	101	no	Terbutylazin D5	x
Thiacloprid	HOKKPVIRMVDYPB-UHFFFAOYSA-N	AS	Insecticide	7	5	15	86	114	114	pt	Clothianidin D5	
Thiamethoxam	NWWZPOKUUAIXIW-DHZHZOJOSA-N	AS	Insecticide	10	8	10	86	104	99	pt	Thiamethoxam D3	
Trifloxystrobin	ONCZDRURRATYFI-TVJDWZFNSA-N	AS	Fungicide	50	10	10	43	90	99	no	Metrafenone D9	x
CT-TP-R417888	JNMMKKYUIIQPDG-UHFFFAOYSA-N	TP	Fungicide TP	45	55	40	76	85	101	no	2,4-D3	
CT-TP-R471811	NLCNUAPJCIAONV-UHFFFAOYSA-N	TP	Fungicide TP	100	15	100	79	67	100	no	2,4-D3	
CT-TP-R611968	IODGSFOOWTXKAE-UHFFFAOYSA-N	TP	Fungicide TP	15	10	5	105	99	98	no	2,4-D3	
CT-TP-SYN507900	WUYYRWIYXBUPBS-UHFFFAOYSA-N	TP	Fungicide TP	50	20	10	68	93	94	no	Fipronil 13C15N2	
Metamitron-desamino	OUSYWCQYMPDAEO-UHFFFAOYSA-N	TP	Herbicide TP	10	10	3	114	113	109	no	Metamitron D5	
Metolachlor-OXA	LNOOSYCKMKZOJB-UHFFFAOYSA-N	TP	Herbicide TP	10	10	10	106	103	104	no	Metolachlor D6	

S4.2 Results

S4.2.1 Hydrological behaviour of inlets

Table S14: Event rainfall needed at different inlets for surface runoff to enter the inlet. The duration of the corresponding rain events equalled 1 to 41 hours (median: 9 hours).

Location	I1	I2	I3	I4
Minimal amount of rainfall leading to discharge (mm)	1.3	1.5	3.6	1.3
Minimal rainfall always leading to discharge (mm)	3.5	11.5	18.8	3.5

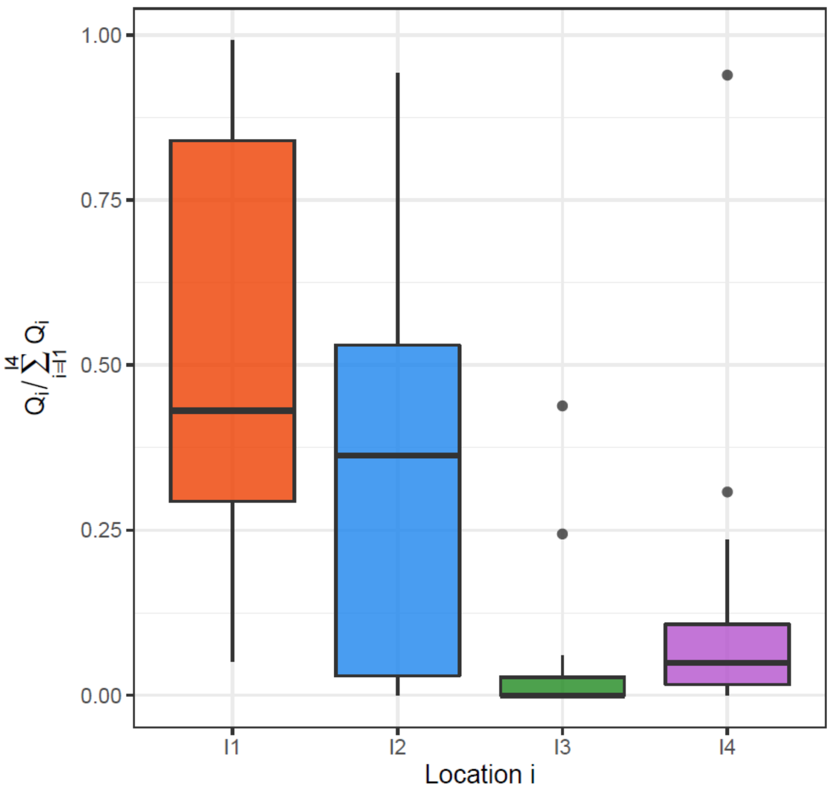


Figure S62: Distribution of the total event discharge ratio between each single inlet and the sum of all four inlets.

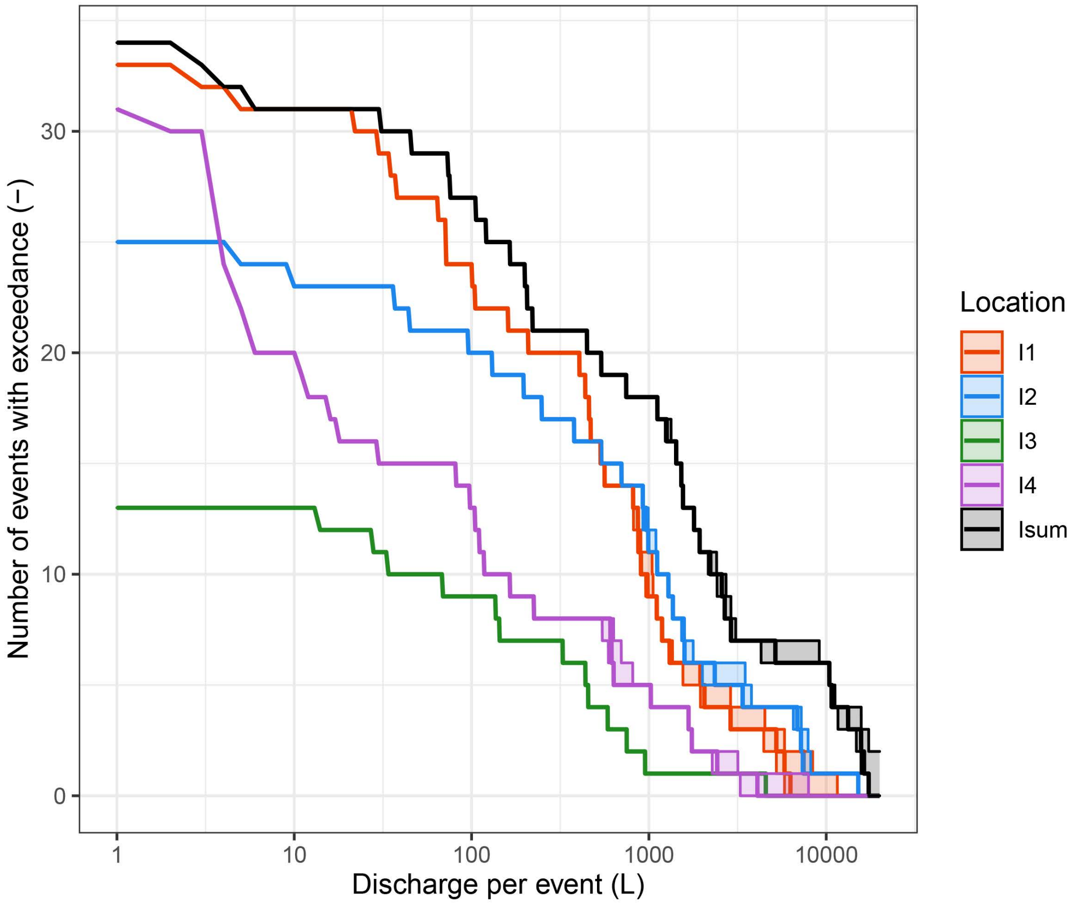


Figure S63: Number of events exceeding a given event discharge in inlets. Isum corresponds to the discharge sum of all four inlets. Bold lines indicate the moderate discharge estimate $Q_{i,e,mod}$. Thin lines indicate the minimum $Q_{i,e,min}$ and high discharge estimates $Q_{i,e,high}$.

Table S15: Fractions of fast and total discharge in the stream originating from inlets for events with total rainfall > 10 mm. Numbers report the moderate estimates. In brackets, the minimum and high estimates are given. The first column shows the measured discharge fractions in the four studied inlets, the second to fourth column show the extrapolation to all inlets in the catchment according to three different methods (i.e. proportional to the road area, the agricultural area, and the number of inlets; see eq. S4.7 to S4.8).

	Measured inlets (I1-I4)	Extrapolation to all inlets		
		Road area	Agri. area	Number of inlets
Fraction of fast discharge $r_{Q,fast}$	0.83% [0.64%; 1.1%]	29% [22%; 38%]	14% [11%; 19%]	33% [25%; 43%]
Fraction of total discharge r_Q	0.22% [0.21%; 0.25%]	7.5% [7.2%; 8.8%]	3.7% [3.6%; 4.4%]	8.5% [8.2%; 10%]

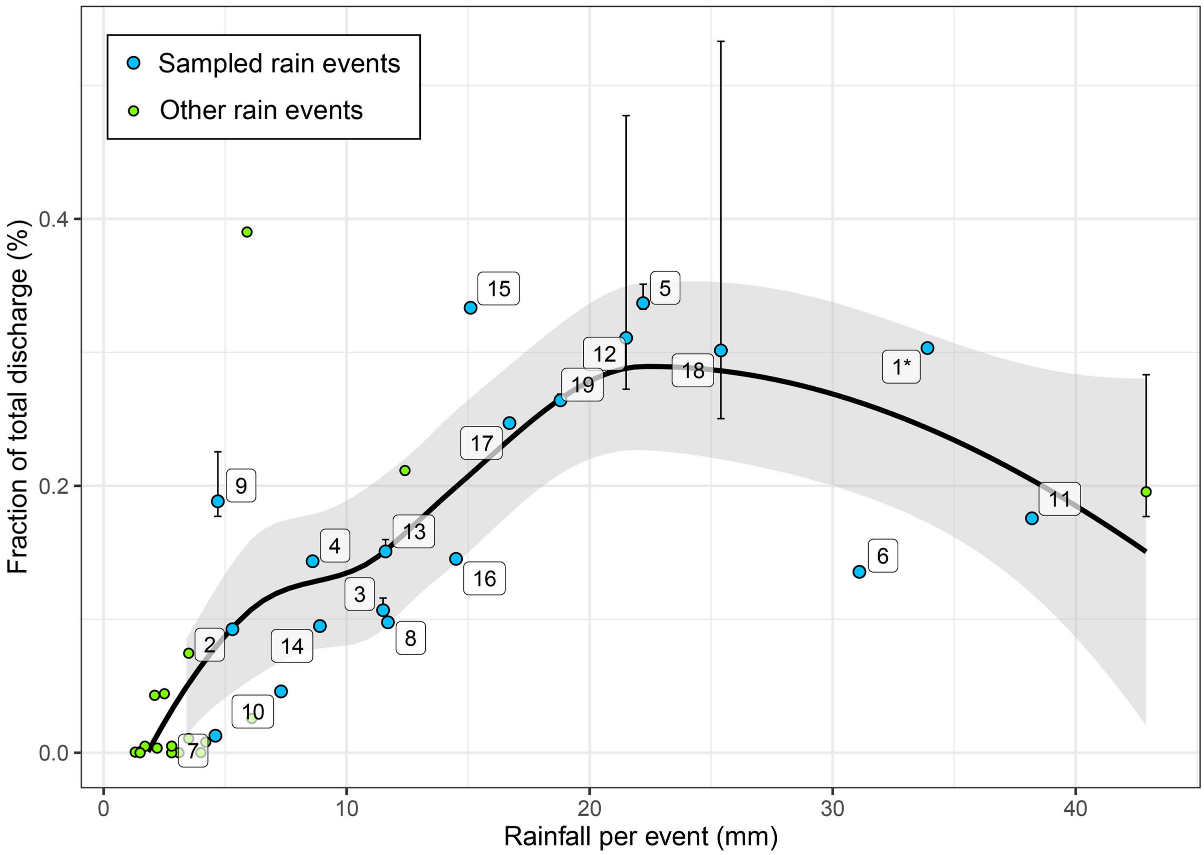


Figure S64: Ratio between total discharge originating from the four inlets and the discharge in the stream r_Q. Points indicate the moderate estimate ($r_{Q,mod}$) and the error bars correspond to the minimum and high estimate ($r_{Q,min}$ and $r_{Q,high}$). Sampling event numbers are indicated with white boxes. The black line represents a smoothed conditional mean (LOESS) of the average r_Q estimates, obtained by a locally weighted scatterplot smoothing (LOESS) using the R package ggplot2 (version 3.3.3, function geom_smooth). The grey area represents the corresponding 95% confidence interval.

S4.2.2 Concentrations and loads

Table S16: Overview over events analysed. I1-I4: Inlets 1 to 4, CS: collector shaft, ST: stream. The column "top ten events" shows the top ten events with respect to sum concentrations in the inlets. The letter A indicates that for the respective event samples in the collector shaft and in the stream were analysed. The letter N indicates that for the respective event only samples in the inlets were analysed. In the column "samples analysed" the sample types are indicated. c: water-level proportional composite sample, g: grab sample after the event, t: time proportional sample, b: time proportional backup sample of cantonal authorities, -: no sample available. Sampling interval: The first number indicates the sampling interval of time proportional samples in the collector shaft that were then pooled together into one composite sample with a total sampling time as indicated in brackets.

Event		Rainfall			Discharge sum (L)					Sum concentrations in I1 to I4 (ng/L)	Top ten events	Samples analysed						Sampling interval CS (min)
ID	Starting time	Sum (mm)	Mean ints. (mm/hr)	Max. ints. (mm/hr)	I1	I2	I3	I4	ST			I1	I2	I3	I4	CS	ST	
1	03.04 13:57	33.9	0.9	6.0	832	8519	162	122	1.4e6	1.7e4		c	c	c	c			
2	26.04 01:57	5.3	0.8	3.6	38	597	0	4	2.8e5	7.1e3		c	g	-	-			
3	27.04 21:42	11.5	0.3	10.8	387	1472	68	99	1.1e6	1.3e4		c	c	c	c			
4	04.05 16:32	8.6	0.5	3.6	555	1329	0	4	5.4e5	5.9e3		c	c	-	-			
5	08.05 13:37	22.2	1.3	6.0	1820	7355	327	1018	2.0e6	4.4e4	A	c	c	c	c	t	t	2 (20)
6	20.05 00:42	31.1	1.4	6.0	2306	1111	3586	1406	2.7e6	2.9e4	A	c	c	c	c	-	t	3 (30)
7	25.05 17:02	4.6	0.5	6.0	139	1	0	2	4.4e5	3.6e4	N	c	g	g	g			
8	28.05 07:17	11.7	1.1	8.4	814	379	439	165	8.8e5	1.7e4		c	c	g	-			
9	29.05 17:57	4.7	1.3	25.2	865	960	136	606	4.3e5	1.9e4	N	c	c	g	c			
10	06.06 10:12	7.3	0.9	4.8	437	5	0	5	9.7e5	1.9e4	A	g	g	g	g	t	b	3 (30)
11	10.06 11:52	38.2	0.9	9.6	5378	7034	951	1721	8.7e6	2.8e4	N	c	c	c	c			
12	15.06 17:57	21.5	8.6	58.8	5230	3379	586	4109	3.5e6	8.9e4	A	c	c	g	c	t	t	3 (30)
13	01.07 18:07	11.6	1.8	27.6	988	249	0	30	7.7e5	6.5e4	A	c	c	c	g	t	b	3 (30)
14	06.07 09:42	8.9	1.8	10.8	642	98	0	9	9.7e5	8.7e4	A	c	g	g	g	-	b	3 (30)
15	14.07 21:47	15.1	2.0	8.4	1185	1547	33	111	7.0e5	2.3e4	N	c	c	g	g			
16	28.07 12:17	14.5	0.8	10.8	427	94	0	12	2.8e5	1.7e4		g	g	g	g			
17	06.08 07:07	16.7	1.1	24.0	865	531	0	117	4.9e5	1.6e4		c	c	g	g			
18	10.08 03:42	25.4	2.0	58.8	2087	2349	144	614	1.6e6	1.3e4		c	c	c	c			
19	18.08 22:32	18.8	2.0	50.4	1311	1099	28	224	8.6e5	1.2e4		g	g	g	g			

Table S17: Overview over the transformation product concentrations measured at the different sampling sites. To calculate mean concentrations, we replaced concentrations below the limit of quantification (LOQ) by zero (lower value reported) and by the LOQ (higher value reported). The transformation product pattern shown here is most likely caused by the low number of transformation products analysed and does not allow for a general conclusion on transport processes involved. I1-I4: inlets, CS: collector shaft, ST: stream.

Site	I1	I2	I3	I4	CS	ST
Mean transformation product concentration (ng L^{-1})	271-301	25-56	36-66	10-43	714-716	1003-1006
Maximal transformation product concentration (ng L^{-1})	7300	870	550	180	5200	5500
Transformation product with highest concentration	Meta-mitron-Desamino	CT-TP-R471811	CT-TP-R471811	Meta-mitron-Desamino	Meta-mitron-Desamino	CT-TP-R471811

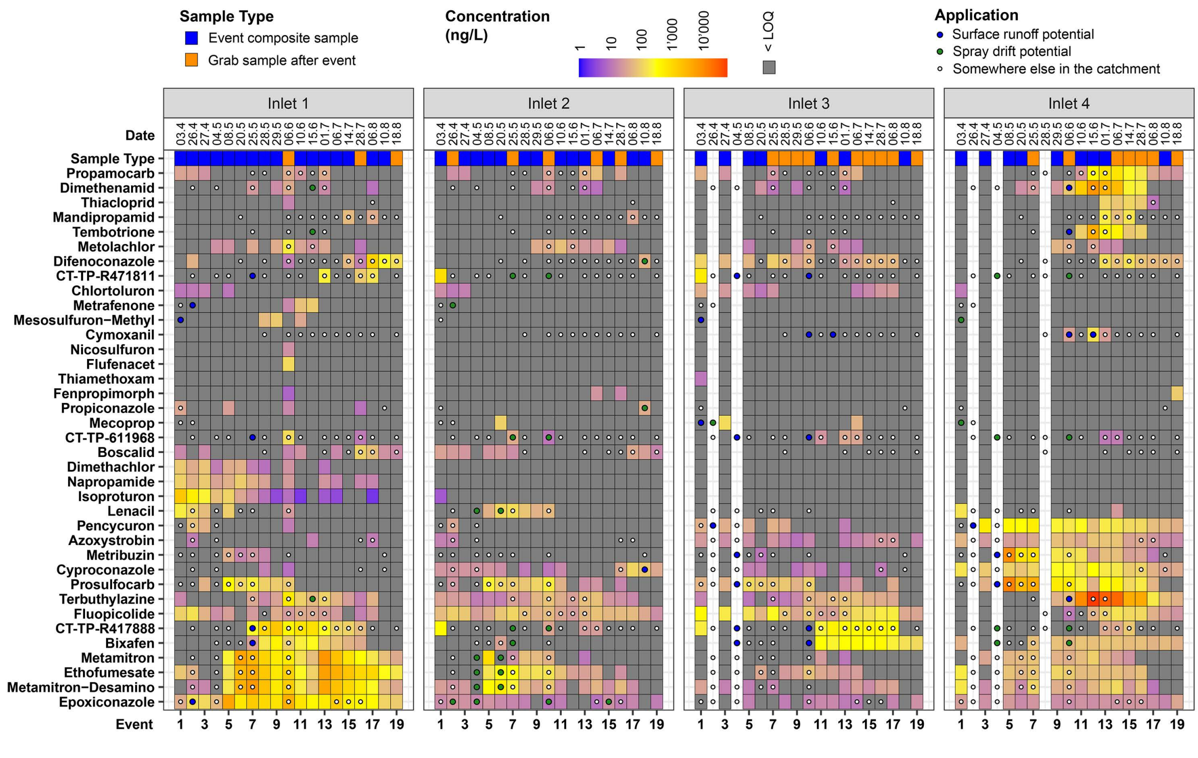


Figure S65: Concentrations c (ng/L) measured in inlets for event 1 (3 April 2019) to 19 (18 August 2019) for all substances that were found in inlets. White rows indicate that no sample was taken. In the first column, the sample type is indicated. In the remaining columns, substances are clustered by the concentrations measured. Coloured dots indicate that the particular substance was applied in the period between the respective and the previous event. Dot colours specify the potential transport processes. LOQ: Limit of quantification.

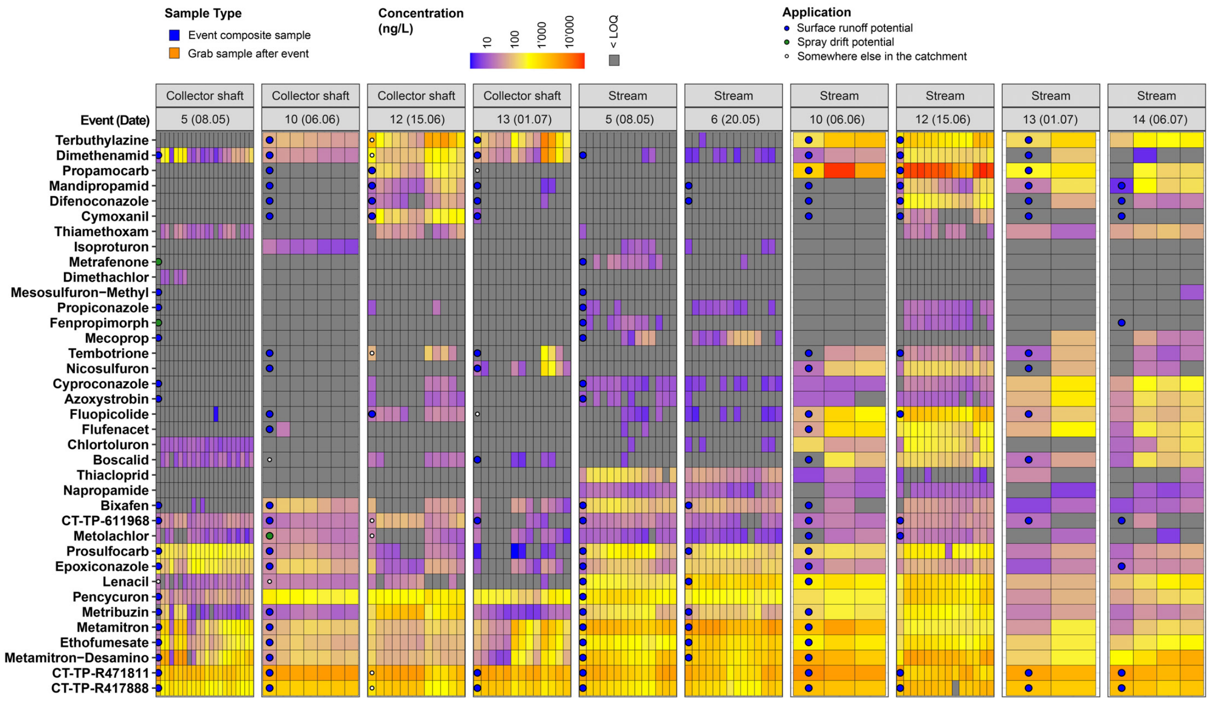


Figure S66: Concentrations c (ng/L) measured in the collector shaft (event 5, 10, 12, and 13) and in the stream (events 5, 6, 10, 12, 13, 14) for all substances found at one of these two sampling sites. Measurements of events 10, 13, and 14 in the stream originate from backup samples of the cantonal authorities. LOQ: Limit of quantification.

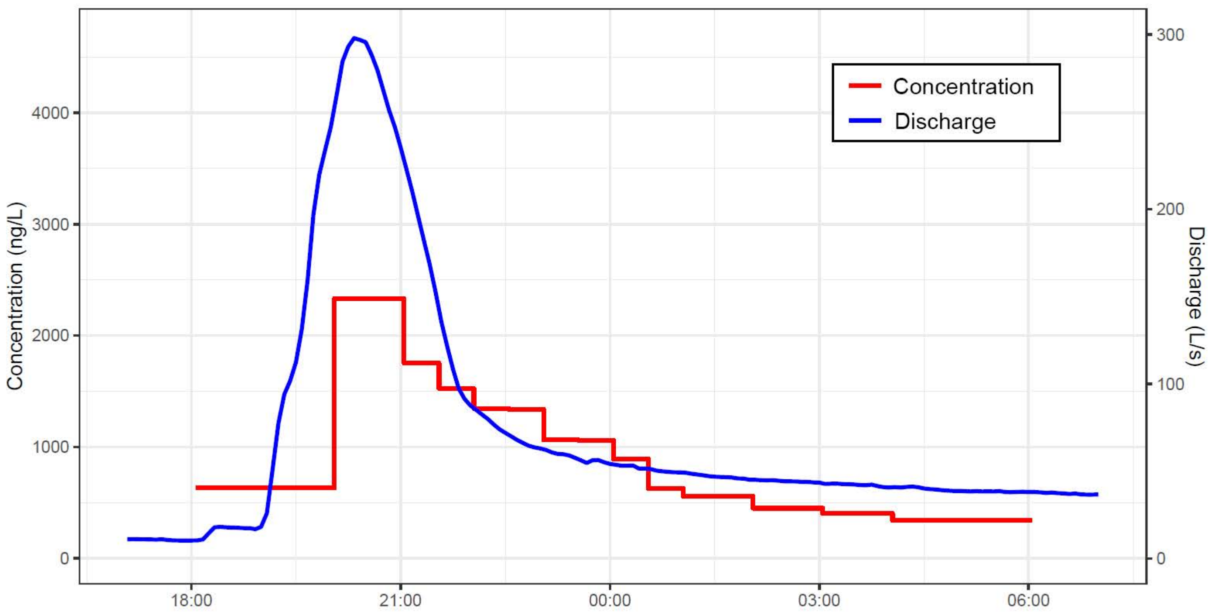


Figure S67: Discharge and terbuthylazine concentration in the stream during event 12 (07.08.2019 to 08.08.2019).

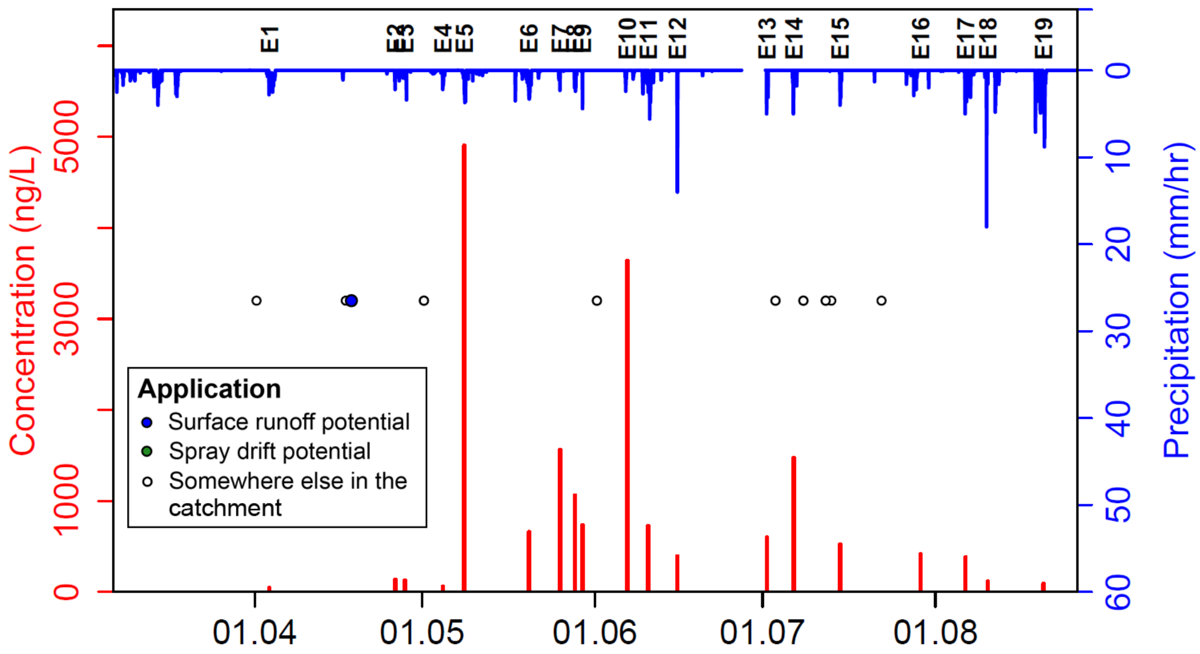


Figure S68: Concentrations of epoxiconazole in inlet 1, during the sampled events (E1 to E19).

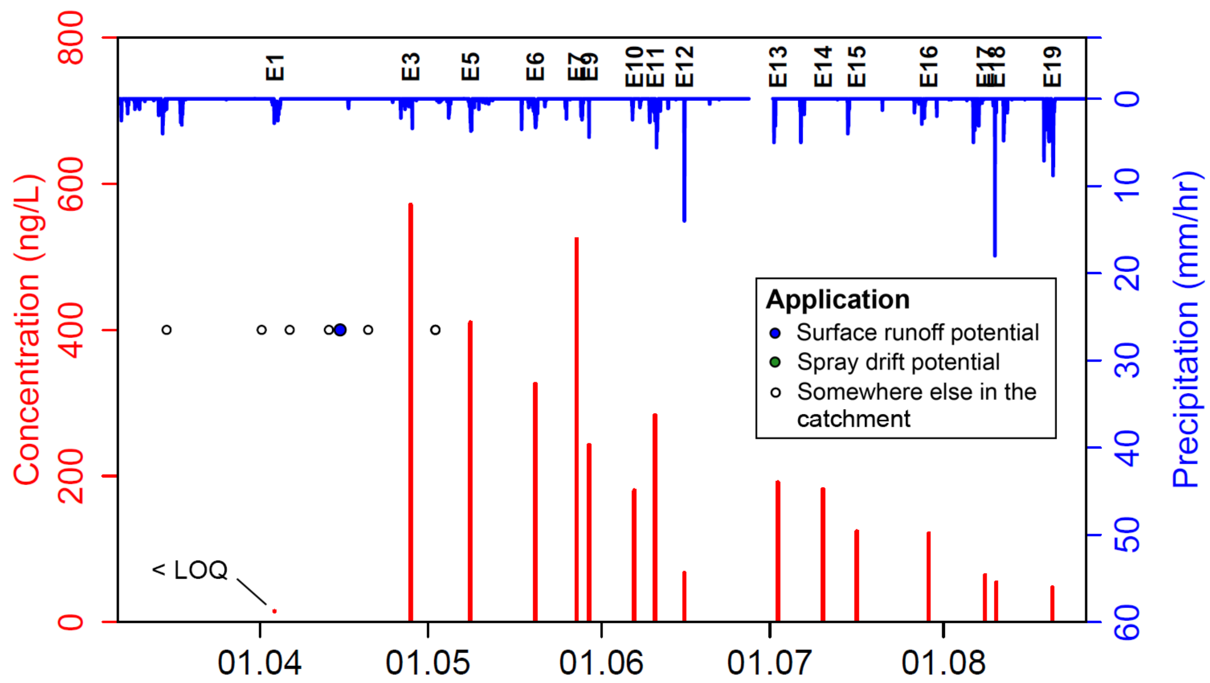


Figure S69: Concentrations of pencycuron in inlet 4, during the sampled events (E1 to E19). During E2, E4, and E8, no samples were taken. No pencycuron was found in the first sample (i.e. the concentration was smaller than the limit of quantification (LOQ) of 15 ng/L).

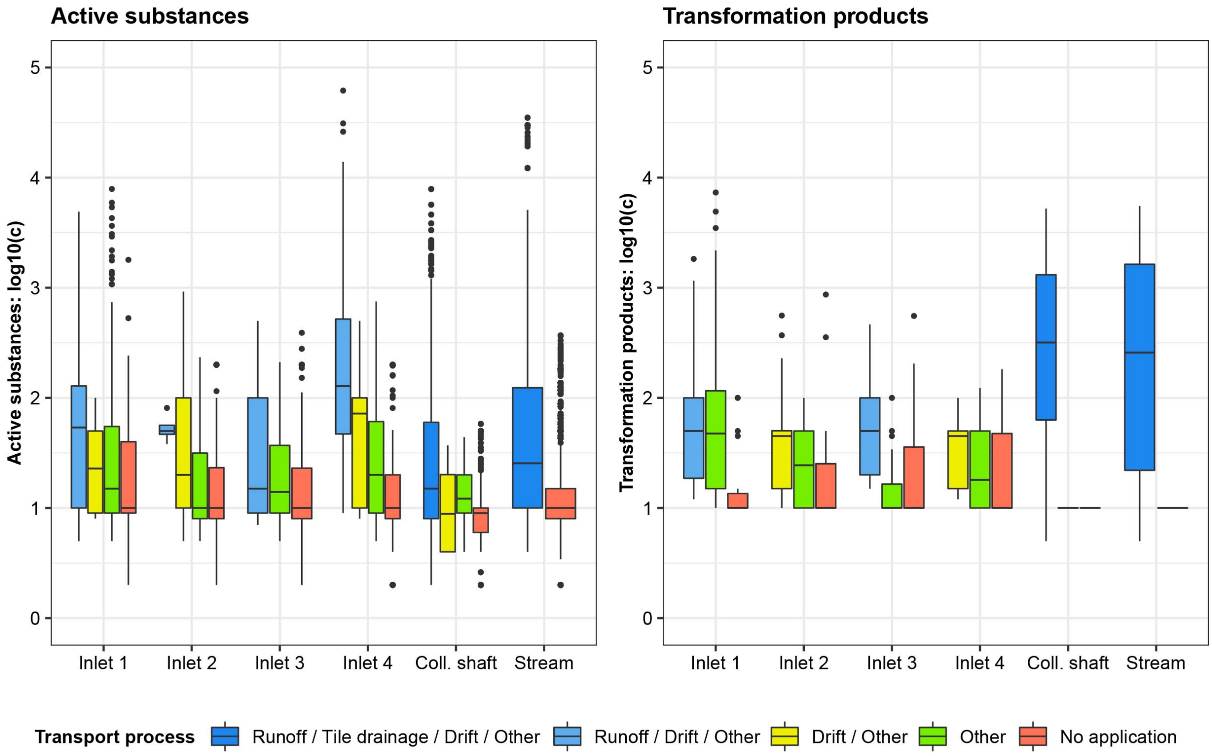


Figure S70: Distribution of pesticide and transformation product concentrations for all sampling sites. Concentrations are assigned to possible responsible transport processes. For substances below the limit of quantification (LOQ), the LOQ was used for the analysis.

Table S18: Result of the linear mixed model. Estimates, t values, and p values are given for each explanatory variable. An explanation of the column "estimates" is provided in the following. For the calculation of p-values, a Kenward-Roger approximation was used for calculating the degrees of freedom. The proportion of variance explained by the fixed factors alone (marginal R2) was 0.25. The proportion explained by the fixed and random factors (conditional R2) equalled 0.48.

Explanatory variable	Abbre-viation	Unit	Estimates: Mean [confidence interval: 2.5% – 97.5%]	t value	p value
Intercept	Int.	-	+ (1.4 [1.0 – 1.9])	6.24	< 0.001
Time since application	t_{appl}	days	– (2.4 [3.9 – 1.0]) · 10^{-3}	-3.35	0.001
Amount of pesticide applied per area (log10)	$\log_{10}(m_{appl})$	g ha^{-1}	+ (2.2 [1.5 – 2.9]) · 10^{-1}	6.24	< 0.001
Freundlich adsorption coefficient normalized to organic carbon content (log10)	$\log_{10}(K_{foc})$	mg L^{-1}	– (2.2 [3.2 – 1.2]) · 10^{-1}	-4.39	< 0.001
Half-life in water	$DT_{50,\ water}$	days	+ (3.2 [1.1 – 5.3]) · 10^{-3}	2.98	0.003
Half-life in soil	$DT_{50,\ soil}$	days	+ (1.1 [0.6 – 1.6]) · 10^{-3}	4.56	< 0.001
Moderate estimate of the discharge in the inlet during the event (log10)	$\log_{10}(Q_{mod})$	L	+ (1.1 [-4.0 – 6.0]) · 10^{-2}	0.45	0.654
Potential transport processes involved	$p_{transport}$	-	– (5.1 [6.3 – 4.0]) · 10^{-1}	-8.54	< 0.001

To improve the understandability of Table S18, the meaning of the "estimates" column is explained in the following. This column represents the mean estimates of the fixed effects of the linear mixed model and their confidence interval (2.5% to 97.5%). These effects corresponds to an intercept for row 1, to a slope for rows 2 to 7, and to a categorical variable effect for row 8.

In the following, the meaning of these estimates is explained on the example of the variable "time since application" (row 2). In a mathematical notation, our mixed model can be written as follows:

$$\log_{10}(c) = \mathrm{Intercept} + m_1 \cdot t_{appl} + m_2 \cdot \log_{10}(m_{appl}) + m_3 \cdot \log_{10}(K_{foc}) + \cdots \qquad \text{(S4.12)}$$

Where m_1, m_2, m_3, … are the estimated slopes, c is the pesticide concentration (ng/L) and t_{appl} is the time since application (days). As shown in Table S18, the mean estimate of the slope m_1 equals $-2.4 \cdot 10^{-3}$ day^{-1}. This means, that the logarithm ($\log_{10}$) of the measured concentration (ng/L) is expected to decrease on average by a factor of $-2.4 \cdot 10^{-3}$ per day after application.

Table S19: Ratio between pesticide loads in inlets and the stream (moderate estimates, eq. S4.7 and S4.8). In square brackets, the minimum and high estimates are given. Columns show the ratios measured for the sampled inlets, and the ratios resulting from extrapolating the measurements to the entire catchment using three different methods, i.e. proportional to the road area, the agricultural area, and the number of inlets.

	Sampled inlets (I1-I4)	Extrapolation to entire catchment		
		Road area	Agri. area	Number of inlets
Mean of single substance load ratios $r_{f,\mu,subst}$	1.8% [0.77%; 3.7%]	61% [27%; 126%]	30% [13%; 64%]	70% [30%; 144%]
Ratio of load sums $r_{f,\mu,sum}$	0.29% [0.24%; 0.52%]	10% [8.5%; 18%]	5.1% [4.2%; 9.1%]	12% [9.7%; 21%]

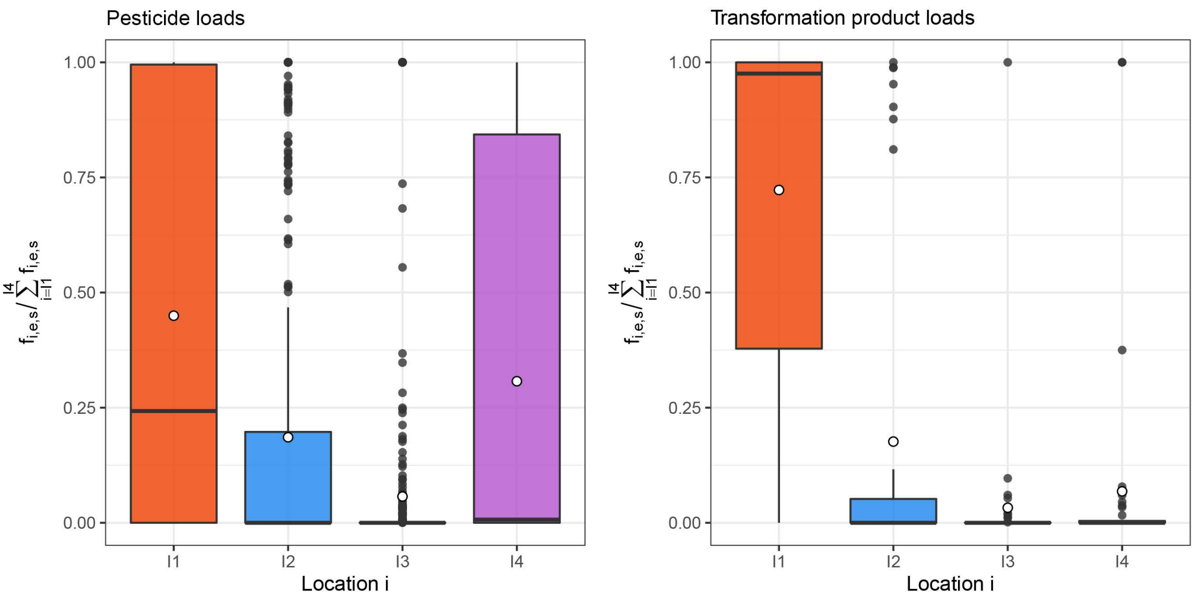


Figure S71: Relative loads per inlet compared to the loads transported through the four measured inlets. Left: Pesticides, right: transformation products. The transformation product pattern shown here is most likely caused by the low number of transformation products analysed and does not allow for general conclusions on the transport processes involved.

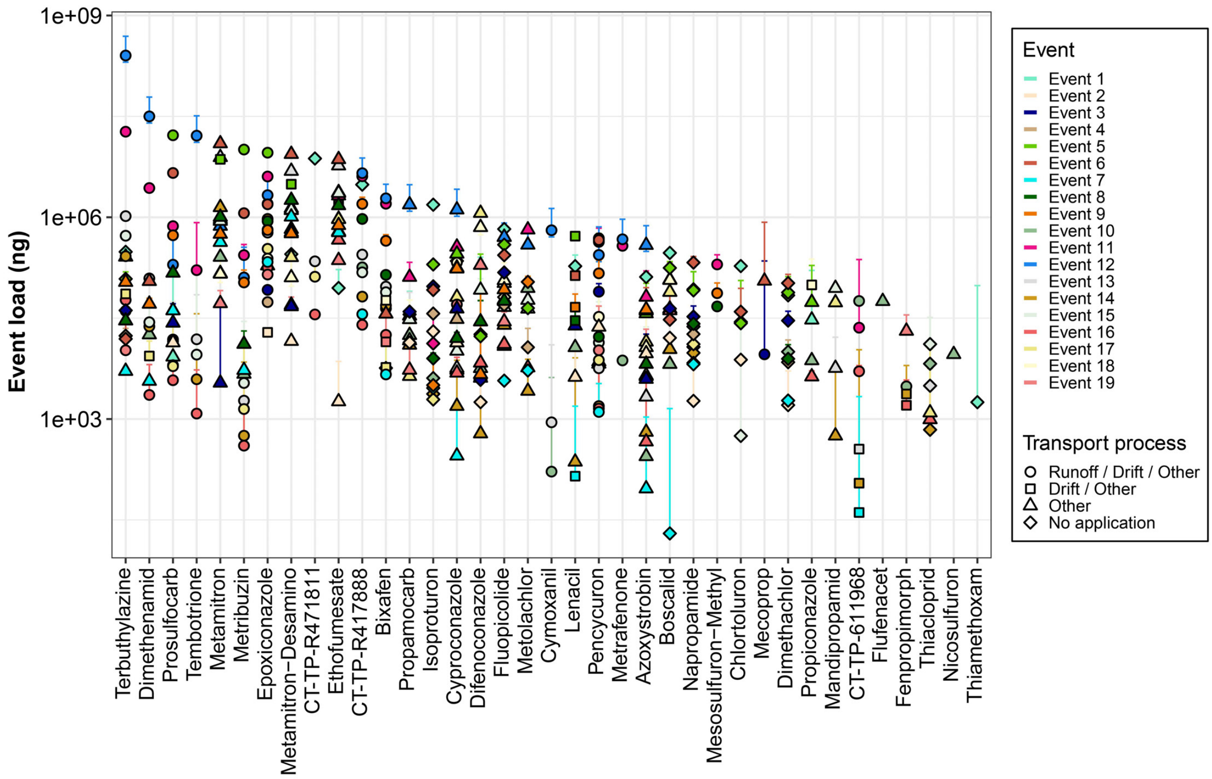


Figure S72: Sum of loads in the four inlets per substance and event. Only loads > 0 ng are shown.

On the following pages, a table with all concentrations measured during the field study is provided (Table S21). Table S20 provides a description for the information contained in each row of Table S21.

Table S20: Metainformation on the concentrations measured during the field study (see following pages).

Row	Description
Event	Sampling event ID
Sampling site	Abbreviation of the sampling site (I1-I4: Inlets 1-4, CS: collector shaft, ST: stream)
Sample type	Type of the sample (WL: water level proportional composite sample, GR: grab sample, TI: time proportional composite sample)
Sampling end	Time point at which sampling ended (DD.MM hh:mm, UTC+1), year 2019
Sampling start	Time point at which sampling started (DD.MM hh:mm, UTC+1), year 2019
Measurement set	Set of chemical analysis during which the sample was measured (set 1, set 2, set 3, set 1&3, or set 2&3)
Azoxystrobin	Concentration of azoxystrobin in (ng/L). If the substance was below LOQ, this is indicated by "<" followed by the LOQ of the respective set.
…	Concentration of other substances in (ng/L). If the substance was below LOQ, this is indicated by "<" followed by the LOQ of the respective set.
Metolachlor-OXA	Concentration of metolachlor-OXA in (ng/L). If the substance was below LOQ, this is indicated by "<" followed by the LOQ of the respective set.

Table S21: Concentrations measured in during the field study. A description of the information contained in each row is provided in Table S20.

See following pages.

Event	1	1	1	1	2	2	3	3	3	3	4	4	5	5
Sampling site	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**ST**	**ST**
Sample type	WL	WL	WL	WL	WL	GR	WL	WL	WL	WL	WL	WL	TI	TI
Sampling end	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	08.05 15:54	08.05 19:24
Sampling start	03.04 15:09	03.04 15:09	03.04 15:09	03.04 15:09	26.04 05:11	26.04 14:30	27.04 22:00	27.04 22:00	27.04 22:00	27.04 22:00	04.05 17:49	04.05 17:49	08.05 13:55	08.05 18:54
Measurement set	1	1	1	1	1	1	1	1	1	1	1	1	2	3
Azoxystrobin	<7	14	26	51	13	15	<7	<7	20	26	<7	<7	11	<6
Bixafen	<8	<8	<8	47	<8	<8	<8	<8	<8	<8	<8	<8	43	146
Boscalid	17	33	<10	<10	<10	27	18	25	<10	<10	<10	<10	<10	6
Carfentrazone-ethyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<20	<15
Chlortoluron	11	20	44	12	12	12	14	14	26	<10	<10	<10	<6	<6
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	<8	24	<8	161	<8	23	<8	24	<8	85	<8	23	<8	7
Difenoconazole	<25	<25	112	<25	47	<25	<25	<25	56	<25	<25	<25	<8	<8
Diflufenican	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<200	<100
Dimethachlor	83	<6	<6	<6	42	<6	75	<6	<6	<6	13	<6	<9	<10
Dimethenamid	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<8	<4
Epoxiconazole	41	18	<7	29	135	24	126	22	<7	21	58	17	15	108
Ethofumesate	78	<9	<9	196	48	<9	<9	<9	<9	<9	<9	<9	189	411
Fenpropimorph	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<5	19
Florasulam	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<50	<20
Fluazifop (free acid)	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<9	<10
Fluazinam	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<8	<4
Flufenacet	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<20	6
Fluopicolide	102	61	390	<8	157	99	49	81	187	<8	20	29	<9	8
Flupyrsulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<9	<10
Foramsulfuron	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<4
Iodosulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<15	<10
Isoproturon	1793	5	<2	<2	529	<2	242	<2	<2	<2	67	<2	<4	8
Lenacil	202	<10	<10	159	111	<10	63	<10	<10	<10	<10	<10	141	225
Mandipropamid	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<9	<4
Mecoprop	<100	<100	<100	<100	<100	<100	<100	<100	134	<100	<100	<100	<40	12
Mesosulfuron-methyl	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<9
Metamitron	<20	<20	<20	<20	<20	<20	<20	<20	<20	35	<20	<20	1305	1765
Metolachlor	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	21	<8	11	8
Metrafenone	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<9	15
Metribuzin	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	71	219
Napropamide	102	<8	<8	<8	49	<8	86	<8	<8	<8	33	<8	11	11
Nicosulfuron	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<8	<6
Pencycuron	<15	<15	44	<15	67	35	49	<15	43	571	<15	<15	282	885
Propamocarb	34	<5	47	<5	31	21	22	20	23	<5	<5	<5	<25	<50
Propiconazole	36	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<8	<6
Prosulfocarb	<15	<15	52	<15	<15	23	49	<15	48	47	<15	<15	44	543
Prothioconazole	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<200	<100
Pyraclostrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<9	<15
Spiroxamine	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<25	<20
Tembotrione	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<8	<8
Terbuthylazine	17	34	15	<9	<9	29	<9	28	<9	<9	<9	13	<7	<6
Thiacloprid	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	33	168
Thiamethoxam	<10	<10	11	<10	<10	<10	<10	<10	<10	<10	<10	<10	10	<10
Trifloxystrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<10	<10
CT-TP-R417888	<45	355	205	<45	<45	<45	<45	<45	<45	<45	<45	<45	1529	642
CT-TP-R471811	<100	867	554	<100	<100	<100	<100	<100	<100	<100	<100	<100	4481	1339
CT-TP-R611968	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	22	31
CT-TP-SYN507900	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	216	199
Metamitron-desamino	<10	30	32	182	18	23	16	26	14	18	<10	<10	1187	652
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	5	5	5	5	5	5	5	5	5	5	5	5	5	5
Sampling site	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**CS**	**CS**
Sample type	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI
Sampling end	08.05 19:54	08.05 20:54	08.05 21:54	08.05 23:54	08.05 16:24	08.05 16:54	08.05 17:14	08.05 17:34	08.05 17:54	08.05 18:14	08.05 18:34	08.05 18:54	08.05 14:19	08.05 17:19
Sampling start	08.05 19:24	08.05 19:54	08.05 20:54	08.05 21:54	08.05 15:54	08.05 16:24	08.05 16:54	08.05 17:14	08.05 17:34	08.05 17:54	08.05 18:14	08.05 18:34	08.05 14:00	08.05 16:59
Measurement set	3	3	3	3	2	2	2	2	2	2	2	2	2	3
Azoxystrobin	6	<6	<6	<6	12	11	10	<9	13	<9	11	12	<9	<6
Bixafen	126	109	97	61	78	87	94	126	140	160	195	198	<20	<5
Boscalid	<4	<4	<4	<4	<10	<10	<10	<10	<10	<10	<10	<10	<10	8
Carfentrazone-ethyl	<15	<15	<15	<15	<20	<20	<20	<20	<20	<20	<20	<20	<20	<15
Chlortoluron	7	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	7
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	9	7	5	<4	9	9	9	11	12	12	12	14	<8	<4
Difenoconazole	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Diflufenican	<100	<100	<100	<100	<200	<200	<200	<200	<200	<200	<200	<200	<200	<100
Dimethachlor	<10	<10	<10	<10	<9	<9	<9	<9	<9	<9	<9	<9	<9	<10
Dimethenamid	<4	<4	6	11	<8	<8	<8	<8	<8	<8	<8	<8	10	8
Epoxiconazole	111	93	76	49	28	22	29	51	54	75	106	106	221	188
Ethofumesate	343	330	250	209	292	319	395	500	511	547	553	384	32	132
Fenpropimorph	19	13	13	<10	<5	<5	<5	8	<5	<5	9	12	<5	<10
Florasulam	<20	<20	<20	<20	<50	<50	<50	<50	<50	<50	<50	<50	<50	<20
Fluazifop (free acid)	<10	<10	<10	<10	<9	<9	<9	<9	<9	<9	<9	<9	<9	<10
Fluazinam	<4	<4	<4	<4	<8	<8	<8	<8	<8	<8	<8	<8	<8	<4
Flufenacet	<6	<6	10	<6	<20	<20	<20	<20	<20	<20	<20	<20	<20	<6
Fluopicolide	7	5	4	<2	11	<9	<9	<9	<9	<9	<9	<9	<9	<2
Flupyrsulfuron-methyl	<10	<10	<10	<10	<9	<9	<9	<9	<9	<9	<9	<9	<9	<10
Foramsulfuron	<4	<4	<4	<4	<9	<9	<9	<9	<9	<9	<9	<9	<9	<4
Iodosulfuron-methyl	<10	<10	<10	<10	<15	<15	<15	<15	<15	<15	<15	<15	<15	<10
Isoproturon	11	14	10	13	<4	<4	<4	<4	<4	<4	<4	<4	<4	<6
Lenacil	174	166	150	146	169	207	220	250	278	260	245	237	19	14
Mandipropamid	<4	<4	<4	<4	<9	<9	<9	<9	<9	<9	<9	<9	<9	<4
Mecoprop	5	37	56	50	<40	<40	<40	<40	<40	<40	<40	<40	<40	<5
Mesosulfuron-methyl	<9	<9	<9	<9	<8	<8	<8	<8	<8	<8	<8	<8	<8	<9
Metamitron	1489	1455	1149	1182	1750	2187	2255	2613	2833	2417	2321	2242	26	208
Metolachlor	9	7	5	<4	11	12	11	12	13	14	14	14	18	8
Metrafenone	19	17	17	8	16	<9	<9	<9	19	<9	27	24	<9	<6
Metribuzin	204	217	261	258	65	63	77	118	126	146	184	192	<7	9
Napropamide	10	10	10	11	15	17	15	18	18	20	18	16	<10	<6
Nicosulfuron	<6	<6	<6	<6	<8	<8	<8	<8	<8	<8	<8	<8	<8	<6
Pencycuron	929	703	663	455	400	353	498	658	633	744	986	1024	19	66
Propamocarb	<50	<50	<50	<50	<25	<25	<25	<25	<25	<25	<25	<25	<25	<50
Propiconazole	<6	<6	<6	<6	<8	<8	<8	9	10	<8	<8	9	<8	<6
Prosulfocarb	676	457	464	343	103	72	79	164	129	221	356	397	173	260
Prothioconazole	<100	<100	<100	<100	<200	<200	<200	<200	<200	<200	<200	<200	<200	<100
Pyraclostrobin	<15	<15	<15	<15	<9	<9	<9	<9	<9	<9	<9	<9	<9	<15
Spiroxamine	<20	<20	<20	<20	<25	<25	<25	<25	<25	<25	<25	<25	<25	<20
Tembotrione	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Terbuthylazine	<6	<6	<6	<6	<7	<7	<7	<7	<7	<7	<7	<7	<7	<6
Thiacloprid	136	119	71	51	68	<5	93	158	180	175	198	178	<5	<15
Thiamethoxam	<10	<10	<10	<10	<8	<8	<8	<8	<8	<8	<8	<8	<8	11
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
CT-TP-R417888	576	506	371	380	1293	1552	1349	1077	1180	1010	856	763	1545	345
CT-TP-R471811	1267	1024	854	996	3234	3916	3981	2879	3085	2533	1946	1941	4652	827
CT-TP-R611968	29	22	29	34	17	16	20	21	19	21	22	24	18	19
CT-TP-SYN507900	169	130	201	230	127	149	133	118	119	123	116	119	462	100
Metamitron-desamino	491	543	422	492	1556	1832	1820	1666	1617	1354	1055	1003	24	229
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	5	5	5	5	5	5	5	5	5	5	5	5	5	5
Sampling site	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**
Sample type	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI
Sampling end	08.05 17:39	08.05 17:59	08.05 18:19	08.05 18:39	08.05 18:59	08.05 19:19	08.05 19:39	08.05 19:59	08.05 20:19	08.05 14:39	08.05 20:39	08.05 20:59	08.05 21:19	08.05 14:59
Sampling start	08.05 17:19	08.05 17:39	08.05 17:59	08.05 18:19	08.05 18:39	08.05 18:59	08.05 19:19	08.05 19:39	08.05 19:59	08.05 14:19	08.05 20:19	08.05 20:39	08.05 20:59	08.05 14:39
Measurement set	2	3	2	3	2	3	2	3	2	3	3	2	3	2
Azoxystrobin	<9	<6	<9	<6	<9	<6	<9	<6	<9	<6	<6	<9	<6	<9
Bixafen	<20	<5	<20	<5	<20	<5	<20	<5	<20	<5	<5	<20	<5	<20
Boscalid	19	7	16	8	23	9	20	10	18	<4	8	18	9	16
Carfentrazone-ethyl	<20	<15	<20	<15	<20	<15	<20	<15	<20	<15	<15	<20	<15	<20
Chlortoluron	<6	10	<6	10	9	9	9	11	12	11	14	<6	11	14
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	<8	<4	<8	<4	<8	<4	<8	<4	<8	<4	<4	<8	<4	<8
Difenoconazole	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Diflufenican	<200	<100	<200	<100	<200	<100	<200	<100	<200	<100	<100	<200	<100	<200
Dimethachlor	<9	<10	<9	<10	<9	<10	<9	10	11	<10	14	11	18	<9
Dimethenamid	26	24	58	58	49	78	144	221	339	5	456	418	525	12
Epoxiconazole	127	121	120	135	88	108	92	89	76	223	55	42	37	195
Ethofumesate	148	196	215	284	167	278	300	485	576	46	899	640	969	37
Fenpropimorph	<5	<10	<5	<10	<5	<10	<5	<10	<5	<10	<10	<5	<10	<5
Florasulam	<50	<20	<50	<20	<50	<20	<50	<20	<50	<20	<20	<50	<20	<50
Fluazifop (free acid)	<9	<10	<9	<10	<9	<10	<9	<10	<9	<10	<10	<9	<10	<9
Fluazinam	<8	<4	<8	<4	<8	<4	<8	<4	<8	<4	<4	<8	<4	<8
Flufenacet	<20	<6	<20	<6	<20	<6	<20	<6	<20	<6	<6	<20	<6	<20
Fluopicolide	<9	<2	<9	<2	<9	<2	<9	<2	<9	<2	<2	<9	<2	<9
Flupyrsulfuron-methyl	<9	<10	<9	<10	<9	<10	<9	<10	<9	<10	<10	<9	<10	<9
Foramsulfuron	<9	<4	<9	<4	<9	<4	<9	<4	<9	<4	<4	<9	<4	<9
Iodosulfuron-methyl	<15	<10	<15	<10	<15	<10	<15	<10	<15	<10	<10	<15	<10	<15
Isoproturon	<4	<6	<4	<6	<4	<6	<4	<6	<4	<6	<6	<4	<6	<4
Lenacil	26	16	25	16	27	14	28	<8	19	<8	14	<15	10	17
Mandipropamid	<9	<4	<9	<4	<9	<4	<9	<4	<9	<4	<4	<9	<4	<9
Mecoprop	<40	<5	<40	<5	<40	<5	<40	<5	<40	<5	<5	<40	<5	<40
Mesosulfuron-methyl	<8	<9	<8	<9	<8	<9	<8	<9	<8	<9	<9	<8	<9	<8
Metamitron	297	287	430	431	345	504	674	996	1298	12	1654	1430	1848	15
Metolachlor	13	6	12	5	12	4	10	<4	10	16	<4	<8	<4	22
Metrafenone	<9	<6	<9	<6	<9	<6	<9	<6	<9	<6	<6	<9	<6	<9
Metribuzin	10	8	<7	6	10	11	34	82	126	6	129	153	161	8
Napropamide	<10	<6	<10	<6	<10	<6	<10	<6	<10	<6	<6	<10	<6	<10
Nicosulfuron	<8	<6	<8	<6	<8	<6	<8	<6	<8	<6	<6	<8	<6	<8
Pencycuron	75	51	48	34	72	68	66	71	59	18	45	41	26	35
Propamocarb	<25	<50	<25	<50	<25	<50	<25	<50	<25	<50	<50	<25	<50	<25
Propiconazole	<8	<6	<8	<6	<8	<6	<8	<6	<8	<6	<6	<8	<6	<8
Prosulfocarb	260	216	234	223	232	213	190	170	189	186	154	123	98	237
Prothioconazole	<200	<100	<200	<100	<200	<100	<200	<100	<200	<100	<100	<200	<100	<200
Pyraclostrobin	<9	<15	<9	<15	<9	<15	<9	<15	<9	<15	<15	<9	<15	<9
Spiroxamine	<25	<20	<25	<20	<25	<20	<25	<20	<25	<20	<20	<25	<20	<25
Tembotrione	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Terbuthylazine	<7	<6	<7	<6	<7	<6	<7	<6	<7	<6	<6	<7	<6	<7
Thiacloprid	<5	<15	<5	<15	<5	<15	<5	<15	<5	<15	<15	<5	<15	<5
Thiamethoxam	26	33	27	<10	13	14	17	14	24	<10	28	34	22	13
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
CT-TP-R417888	599	766	786	470	321	380	353	411	509	959	591	711	858	538
CT-TP-R471811	1537	1626	1800	1162	779	905	794	890	1090	2658	1322	1508	1927	2246
CT-TP-R611968	29	41	36	35	25	29	33	36	35	21	47	40	50	14
CT-TP-SYN507900	154	207	168	130	102	133	108	131	149	282	222	185	281	152
Metamitron-desamino	346	418	777	937	652	1157	1744	2740	3415	7	4541	3781	5237	<10
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	**5**	**5**	**5**	**5**	**5**	**5**	**5**	**5**	**5**	**5**	**6**	**6**	**6**	**6**
Sampling site	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**I1**	**I2**	**I3**	**I4**	**ST**	**ST**	**ST**	**ST**
Sample type	TI	TI	TI	TI	TI	TI	WL	WL	WL	WL	TI	TI	TI	TI
Sampling end	08.05 15:19	08.05 15:39	08.05 15:59	08.05 16:19	08.05 16:39	08.05 16:59	NA	NA	NA	NA	20.05 04:29	20.05 07:59	20.05 08:29	20.05 09:29
Sampling start	08.05 14:59	08.05 15:19	08.05 15:39	08.05 15:59	08.05 16:19	08.05 16:39	08.05 13:52	08.05 13:52	08.05 13:52	08.05 13:52	20.05 02:29	20.05 07:29	20.05 07:59	20.05 08:29
Measurement set	3	2	3	2	3	2	1	1	1	1&3	3	3	3	3
Azoxystrobin	<6	<9	<6	<9	<6	<9	<7	<7	20	30	6	<6	<6	<6
Bixafen	7	<20	7	<20	<5	<20	<8	<8	<8	<8	40	54	54	54
Boscalid	10	15	9	17	10	19	<10	24	<10	<10	<4	<4	<4	<4
Carfentrazone-ethyl	<15	<20	<15	<20	<15	<20	<10	<10	<10	<15	<15	<15	<15	<15
Chlortoluron	14	11	13	12	10	7	11	<10	20	<10	6	<6	<6	<6
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	<4	<8	<4	<8	<4	<8	<8	24	14	103	5	<4	6	6
Difenoconazole	<8	<8	<8	<8	<8	<8	<25	<25	52	<25	<8	<8	<8	<8
Diflufenican	<100	<200	<100	<200	<100	<200	<500	<500	<500	<500	<100	<100	<100	<100
Dimethachlor	<10	<9	<10	<9	<10	<9	42	<6	<6	<10	<10	<10	<10	<10
Dimethenamid	8	18	6	11	5	11	<9	<9	<9	<9	4	<4	4	<4
Epoxiconazole	321	360	287	233	244	175	4903	23	14	27	123	100	104	88
Ethofumesate	47	64	61	44	71	85	161	160	<9	73	1450	707	738	629
Fenpropimorph	<10	<5	<10	<5	<10	<5	<6	<6	<6	<10	<10	<10	<10	<10
Florasulam	<20	<50	<20	<50	<20	<50	<100	<100	<100	<100	<20	<20	<20	<20
Fluazifop (free acid)	<10	<9	<10	<9	<10	<9	<40	<40	<40	<40	57	131	113	83
Fluazinam	<4	<8	<4	<8	<4	<8	<200	<200	<200	<200	<4	<4	<4	<4
Flufenacet	<6	<20	<6	<20	<6	<20	<8	<8	<8	<8	<6	<6	<6	<6
Fluopicolide	<2	<9	<2	<9	3	<9	30	39	152	<8	5	<2	5	<2
Flupyrsulfuron-methyl	<10	<9	<10	<9	<10	<9	<10	<10	<10	<10	<10	<10	<10	<10
Foramsulfuron	<4	<9	<4	<9	<4	<9	<9	<9	<9	<9	<4	<4	<4	<4
Iodosulfuron-methyl	<10	<15	<10	<15	<10	<15	<10	<10	<10	<10	<10	<10	<10	<10
Isoproturon	<6	<4	<6	<4	<6	<4	109	<2	<2	<6	7	<6	<6	<6
Lenacil	12	19	13	20	12	24	98	47	<10	<10	2330	702	905	952
Mandipropamid	<4	<9	<4	<9	<4	<9	<8	<8	<8	<8	<4	<4	<4	<4
Mecoprop	<5	<40	<5	<40	<5	<40	<100	<100	<100	<100	<5	50	75	79
Mesosulfuron-methyl	<9	<8	<9	<8	<9	<8	<8	<8	<8	<9	<9	<9	<9	<9
Metamitron	16	87	67	80	109	204	241	921	<20	53	5095	1963	2006	1874
Metolachlor	14	17	12	16	9	14	22	<8	14	<8	6	7	7	5
Metrafenone	<6	<9	<6	<9	<6	<9	<15	<15	<15	<15	<6	<6	<6	9
Metribuzin	6	<7	6	<7	9	11	44	<9	<9	9906	30	101	94	98
Napropamide	<6	<10	<6	<10	<6	<10	45	<8	<8	<8	16	7	9	8
Nicosulfuron	<6	<8	<6	<8	<6	<8	<10	<10	<10	<10	<6	<6	<6	<6
Pencycuron	19	30	26	38	45	75	<15	<15	40	410	205	288	291	330
Propamocarb	<50	<25	<50	<25	<50	<25	<5	<5	<5	<50	<50	<50	<50	<50
Propiconazole	<6	<8	<6	<8	<6	<8	30	<15	<15	<15	<6	9	6	7
Prosulfocarb	289	243	244	295	260	334	348	234	125	13918	58	224	244	214
Prothioconazole	<100	<200	<100	<200	<100	<200	<100	<100	<100	<100	<100	<100	<100	<100
Pyraclostrobin	<15	<9	<15	<9	<15	<9	<50	<50	<50	<50	<15	<15	<15	<15
Spiroxamine	<20	<25	<20	<25	<20	<25	<100	<100	<100	<100	<20	<20	<20	<20
Tembotrione	<8	<8	<8	<8	<8	<8	<50	<50	<50	<50	<8	<8	<8	<8
Terbuthylazine	<6	<7	<6	<7	<6	<7	<9	16	11	<9	<6	<6	<6	<6
Thiacloprid	<15	<5	<15	<5	<15	<5	<7	<7	<7	<15	33	31	32	26
Thiamethoxam	11	16	16	13	<10	12	<10	<10	<10	<10	<10	<10	<10	<10
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<50	<50	<50	<50	<10	<10	<10	<10
CT-TP-R417888	655	693	708	367	446	279	<45	<45	<45	<45	1548	203	376	344
CT-TP-R471811	1830	2153	1863	1509	1199	1113	<100	<100	<100	<100	3762	444	762	683
CT-TP-R611968	16	22	24	19	20	19	<15	<15	<15	<15	20	10	9	<5
CT-TP-SYN507900	178	169	218	102	129	97	<50	<50	<50	<50	261	48	65	61
Metamitron-desamino	14	122	78	91	125	224	180	370	13	50	1514	601	517	575
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	6	6	6	6	6	6	6	6	6	6	6	6	6	6
Sampling site	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**I1**	**I2**	**I3**	**I4**
Sample type	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	WL	WL	WL	WL
Sampling end	20.05 10:29	20.05 12:29	20.05 04:59	20.05 05:29	20.05 05:49	20.05 06:09	20.05 06:29	20.05 06:49	20.05 07:09	20.05 07:29	NA	NA	NA	NA
Sampling start	20.05 09:29	20.05 10:29	20.05 04:29	20.05 04:59	20.05 05:29	20.05 05:49	20.05 06:09	20.05 06:29	20.05 06:49	20.05 07:09	20.05 03:45	20.05 03:45	20.05 03:45	20.05 03:45
Measurement set	3	3	3	3	3	3	3	3	3	3	1	1	1	1
Azoxystrobin	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<7	<7	<7	29
Bixafen	42	34	43	39	61	54	72	80	84	74	<8	33	<8	<8
Boscalid	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<10	27	<10	<10
Carfentrazone-ethyl	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<10	<10	<10	<10
Chlortoluron	<6	<6	7	6	<6	<6	6	<6	<6	<6	<10	<10	11	<10
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	<4	<4	5	8	5	7	6	<4	<4	7	<8	20	14	75
Difenoconazole	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<25	<25	<25	<25
Diflufenican	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<500	<500	<500	<500
Dimethachlor	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	45	<6	<6	<6
Dimethenamid	<4	7	5	5	<4	5	<4	<4	7	6	<9	<9	23	21
Epoxiconazole	81	70	128	128	113	109	113	115	115	111	657	18	<7	18
Ethofumesate	862	1120	925	1423	1128	1206	1344	1605	1560	1128	2917	300	37	53
Fenpropimorph	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<6	<6	<6	<6
Florasulam	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<100	<100	<100	<100
Fluazifop (free acid)	67	54	125	177	196	223	243	238	198	179	<40	<40	<40	<40
Fluazinam	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<200	<200	<200	<200
Flufenacet	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<8	<8	<8	<8
Fluopicolide	<2	<2	4	5	6	3	<2	<2	<2	6	<8	78	52	<8
Flupyrsulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Foramsulfuron	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<9	<9	<9	<9
Iodosulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Isoproturon	<6	<6	6	8	<6	<6	<6	<6	<6	<6	36	<2	<2	<2
Lenacil	1157	1435	993	1331	885	782	918	1091	1436	902	<10	122	<10	<10
Mandipropamid	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<8	<8	<8	<8
Mecoprop	78	53	<5	<5	7	14	8	10	7	14	<100	102	<100	<100
Mesosulfuron-methyl	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<8	<8	<8	<8
Metamitron	1763	1753	3232	4859	3554	3696	4440	4385	4639	3078	5381	<20	<20	58
Metolachlor	<4	8	7	7	5	6	7	11	9	7	<8	<8	<8	<8
Metrafenone	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<15	<15	<15	<15
Metribuzin	158	208	52	48	55	72	104	113	107	93	16	<9	11	765
Napropamide	6	<6	14	18	18	16	18	14	13	13	92	<8	<8	<8
Nicosulfuron	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<10	<10	<10	<10
Pencycuron	273	210	273	247	294	315	322	328	304	307	<15	<15	<15	326
Propamocarb	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<5	<5	<5	<5
Propiconazole	<6	<6	<6	6	<6	6	9	8	9	11	<15	<15	<15	<15
Prosulfocarb	191	139	114	78	123	135	137	197	201	231	120	113	87	2736
Prothioconazole	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Pyraclostrobin	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<50	<50	<50	<50
Spiroxamine	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<100	<100	<100	<100
Tembotrione	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<50	<50	<50	<50
Terbuthylazine	<6	<6	<6	<6	<6	<6	6	<6	<6	<6	<9	14	<9	<9
Thiacloprid	20	19	36	55	53	52	58	50	45	39	<7	<7	<7	<7
Thiamethoxam	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<50	<50	<50	<50
CT-TP-R417888	387	374	1347	1489	1074	1022	951	854	817	715	<45	<45	<45	<45
CT-TP-R471811	850	903	3459	3560	2466	2242	2169	1939	1872	1578	<100	<100	<100	<100
CT-TP-R611968	<5	<5	<5	18	14	15	15	14	16	12	<15	<15	<15	<15
CT-TP-SYN507900	90	137	196	216	152	139	155	123	126	99	<50	<50	<50	<50
Metamitron-desamino	934	1089	1230	1900	1446	1406	1649	1411	1260	894	3486	559	<10	18
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	7	7	7	7	8	8	8	9	9	9	9	10	10	10
Sampling site	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**I3**	**I1**	**I2**	**I3**	**I4**	**ST**	**ST**	**ST**
Sample type	WL	GR	GR	GR	WL	WL	GR	WL	WL	GR	WL	TI	TI	TI
Sampling end	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	06.06 16:00	07.06 00:00	07.06 08:00
Sampling start	25.05 17:26	27.05 13:00	27.05 13:00	27.05 13:00	28.05 09:05	28.05 09:05	29.05 11:50	29.05 18:03	29.05 18:03	29.05 18:03	29.05 18:03	06.06 08:00	06.06 16:00	07.06 00:00
Measurement set	1&3	1	1	1	1	1	1	1	1	1	1	2	2&3	2
Azoxystrobin	<7	<7	24	38	<7	<7	15	<7	<7	10	68	12	12	<9
Bixafen	32	<8	<8	68	171	<8	<8	468	<8	<8	69	<20	23	<20
Boscalid	<10	20	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	192	77
Carfentrazone-ethyl	<15	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<20	<20	<20
Chlortoluron	<10	<10	22	<10	<10	<10	<10	<10	<10	<10	<10	137	43	45
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	<8	<8	<8	117	14	12	<8	<8	<8	10	281	13	11	12
Difenoconazole	<25	<25	94	<25	<25	<25	64	<25	<25	34	<25	<8	<8	<8
Diflufenican	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<200	<200	<200
Dimethachlor	14	<6	<6	<6	10	<6	<6	<6	<6	<6	<6	<9	<10	<9
Dimethenamid	26	<9	11	27	<9	<9	<9	19	19	12	25	15	34	39
Epoxiconazole	1561	16	<7	24	1059	14	13	732	<7	<7	20	16	39	30
Ethofumesate	4294	309	<9	82	1768	129	27	633	150	32	108	450	910	549
Fenpropimorph	<10	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<5	<10	<5
Florasulam	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<50	<50	<50
Fluazifop (free acid)	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	126	50	65
Fluazinam	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<8	<8	<8
Flufenacet	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	48	814	760
Fluopicolide	26	115	279	<8	<8	67	72	42	45	36	<8	66	943	395
Flupyrsulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<9	<10	<9
Foramsulfuron	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	64	25
Iodosulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<15	<15	<15
Isoproturon	21	<2	<2	<2	10	<2	<2	4	<2	<2	<2	<4	<6	<4
Lenacil	<10	142	<10	<10	<10	77	<10	<10	48	<10	<10	514	756	518
Mandipropamid	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<9	<9	<9
Mecoprop	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<40	<40	<40
Mesosulfuron-methyl	<9	<8	<8	<8	58	<8	<8	86	<8	<8	<8	<8	<9	<8
Metamitron	3069	21	<20	72	1209	66	<20	554	58	<20	49	1678	3895	1967
Metolachlor	38	<8	<8	<8	<8	<8	<8	42	42	18	50	<8	17	25
Metrafenone	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<9	<9	<9
Metribuzin	23	<9	<9	882	16	<9	<9	<9	<9	<9	177	132	239	185
Napropamide	47	<8	<8	<8	32	<8	<8	<8	<8	<8	<8	<10	16	11
Nicosulfuron	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	18	101	125
Pencycuron	<15	<15	48	524	<15	<15	38	<15	<15	<15	242	164	251	239
Propamocarb	<50	<5	23	<5	<5	<5	<5	<5	<5	<5	<5	1153	29900	3428
Propiconazole	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<8	<8	<8
Prosulfocarb	222	89	76	4170	103	78	81	90	111	49	575	123	170	136
Prothioconazole	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<200	<200	<200
Pyraclostrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<9	<15	<9
Spiroxamine	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<25	<25	<25
Tembotrione	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<8	37	46
Terbuthylazine	37	31	13	<9	18	24	12	32	53	17	46	165	1436	1746
Thiacloprid	<15	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	7	21	11
Thiamethoxam	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<8	<10	<8
Trifloxystrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<10	<10	<10
CT-TP-R417888	261	<45	<45	<45	1159	<45	<45	1827	<45	<45	<45	1772	1633	1393
CT-TP-R471811	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	5530	4087	3993
CT-TP-R611968	<15	41	<15	<15	<15	<15	<15	<15	<15	<15	<15	17	23	17
CT-TP-SYN507900	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	258	175	209
Metamitron-desamino	7327	229	15	108	2184	58	<10	592	37	<10	43	2795	2220	1307
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	10	10	10	10	10	10	10	10	10	10	10	11	11	11
Sampling site	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**I3**
Sample type	TI	TI	TI	TI	TI	TI	TI	GR	GR	GR	GR	WL	WL	WL
Sampling end	06.06 12:50	06.06 13:20	06.06 13:50	06.06 14:20	06.06 14:50	06.06 15:20	06.06 15:50	NA	NA	NA	NA	NA	NA	NA
Sampling start	06.06 12:21	06.06 12:50	06.06 13:20	06.06 13:50	06.06 14:20	06.06 14:50	06.06 15:20	06.06 18:55	06.06 20:22	06.06 20:12	06.06 19:52	10.06 12:19	10.06 12:19	10.06 12:19
Measurement set	2	2	2	2	2	2	2	2	2	1	1	1	1	1
Azoxystrobin	<9	<9	<9	<9	<9	<9	<9	<9	<9	16	54	<7	<7	<7
Bixafen	67	104	100	95	60	35	38	100	<20	<8	52	217	<8	314
Boscalid	<10	<10	<10	<10	<10	<10	<10	15	<10	<10	<10	22	<10	<10
Carfentrazone-ethyl	<20	<20	<20	<20	<20	<20	<20	<20	<20	<10	<10	<10	<10	<10
Chlortoluron	<6	<6	<6	<6	<6	<6	<6	<6	<6	<10	<10	<10	<10	<10
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	32	<9	<9	<9
Cyproconazole	<8	<8	<8	<8	<8	<8	<8	9	11	16	268	<8	17	12
Difenoconazole	<8	<8	<8	<8	<8	<8	<8	13	<8	53	<25	<25	<25	<25
Diflufenican	<200	<200	<200	<200	<200	<200	<200	<200	<200	<500	<500	<500	<500	<500
Dimethachlor	<9	<9	<9	<9	<9	<9	<9	23	<9	<6	<6	<6	<6	<6
Dimethenamid	45	39	37	35	24	23	22	40	22	<9	80	<9	<9	<9
Epoxiconazole	31	113	135	125	98	53	53	3636	11	14	19	724	13	17
Ethofumesate	63	84	113	87	80	60	60	1396	163	69	87	273	70	38
Fenpropimorph	<5	<5	<5	<5	<5	<5	<5	7	<5	<6	<6	<6	<6	<6
Florasulam	<50	<50	<50	<50	<50	<50	<50	<50	<50	<100	<100	<100	<100	<100
Fluazifop (free acid)	<9	<9	<9	<9	<9	<9	<9	<9	<9	<40	<40	<40	<40	<40
Fluazinam	<8	<8	<8	<8	<8	<8	<8	<8	<8	<200	<200	<200	<200	<200
Flufenacet	<20	21	<20	<20	<20	<20	<20	129	<20	<8	<8	<8	<8	<8
Fluopicolide	<9	<9	<9	<9	<9	<9	<9	27	61	83	12	41	36	33
Flupyrsulfuron-methyl	<9	<9	<9	<9	<9	<9	<9	<9	<9	<10	<10	<10	<10	<10
Foramsulfuron	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Iodosulfuron-methyl	<15	<15	<15	<15	<15	<15	<15	<15	<15	<10	<10	<10	<10	<10
Isoproturon	21	14	11	12	8	7	6	9	<4	<2	<2	2	<2	<2
Lenacil	26	24	23	23	20	18	18	26	58	<10	<10	<10	<10	<10
Mandipropamid	<9	<9	<9	<9	<9	<9	<9	<9	<9	<8	<8	<8	<8	<8
Mecoprop	<40	<40	<40	<40	<40	<40	<40	<40	<40	<100	<100	<100	<100	<100
Mesosulfuron-methyl	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	37	<8	<8
Metamitron	52	73	113	137	120	97	99	585	47	<20	44	150	<20	26
Metolachlor	37	30	33	31	22	19	20	201	43	39	44	34	67	<8
Metrafenone	<9	<9	<9	<9	<9	<9	<9	17	<9	<15	<15	70	<15	<15
Metribuzin	16	14	19	23	16	12	14	<7	<7	<9	144	<9	<9	<9
Napropamide	<10	<10	<10	<10	<10	<10	<10	27	<10	<8	<8	<8	<8	<8
Nicosulfuron	<8	<8	<8	<8	<8	<8	<8	21	<8	<10	<10	<10	<10	<10
Pencycuron	503	339	348	295	240	235	277	13	<9	<15	180	<15	<15	<15
Propamocarb	<25	<25	<25	<25	<25	<25	<25	40	32	<5	<5	24	<5	<5
Propiconazole	<8	<8	<8	<8	<8	<8	<8	17	<8	<15	<15	<15	<15	<15
Prosulfocarb	31	36	48	55	36	26	25	85	210	101	611	<15	46	<15
Prothioconazole	<200	<200	<200	<200	<200	<200	<200	<200	<200	<100	<100	<100	<100	<100
Pyraclostrobin	<9	<9	<9	<9	<9	<9	<9	<9	<9	<50	<50	<50	<50	<50
Spiroxamine	<25	<25	<25	<25	<25	<25	<25	<25	<25	<100	<100	<100	<100	<100
Tembotrione	<8	<8	<8	<8	<8	<8	<8	<8	<8	<50	<50	<50	<50	<50
Terbuthylazine	61	76	72	59	50	41	42	589	72	52	60	102	42	44
Thiacloprid	<5	<5	<5	<5	<5	<5	<5	15	<5	<7	<7	<7	<7	<7
Thiamethoxam	<8	<8	<8	<8	<8	<8	<8	<8	<8	<10	<10	<10	<10	<10
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<10	<10	<50	<50	<50	<50	<50
CT-TP-R417888	1159	1162	1130	1171	1301	1446	1378	412	<55	<45	<45	716	<45	229
CT-TP-R471811	3440	3790	3450	3585	4324	4014	4372	<15	<15	<100	<100	<100	<100	<100
CT-TP-R611968	17	19	24	23	22	22	21	130	11	<15	<15	<15	<15	24
CT-TP-SYN507900	298	290	289	275	373	358	352	<20	<20	<50	<50	<50	<50	<50
Metamitron-desamino	41	82	125	122	116	86	98	1524	85	<10	48	201	21	<10
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	11	12	12	12	12	12	12	12	12	12	12	12
Sampling site	**I4**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**	**ST**
Sample type	WL	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI
Sampling end	NA	15.06 20:03	16.06 01:03	16.06 02:03	16.06 03:03	16.06 04:03	16.06 06:03	15.06 21:03	15.06 21:33	15.06 22:03	15.06 22:33	15.06 23:03
Sampling start	10.06 12:19	15.06 18:04	16.06 00:33	16.06 01:03	16.06 02:03	16.06 03:03	16.06 04:03	15.06 20:03	15.06 21:03	15.06 21:33	15.06 22:03	15.06 22:33
Measurement set	1&3	2	2&3	2	2	2	2	2	2	2	2	2
Azoxystrobin	38	12	13	12	12	<9	12	<9	14	16	15	15
Bixafen	72	<20	35	37	30	26	24	51	41	49	52	48
Boscalid	<10	95	94	112	83	66	67	195	152	149	143	143
Carfentrazone-ethyl	<15	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20
Chlortoluron	<10	18	171	182	177	129	84	51	267	307	288	281
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	47	72	55	49	22
Cyproconazole	135	<8	18	19	16	13	10	17	25	26	30	25
Difenoconazole	<25	43	160	146	118	81	74	165	281	283	311	272
Diflufenican	<500	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Dimethachlor	<10	<9	<10	<9	<9	<9	<9	<9	<9	<9	<9	<9
Dimethenamid	1581	31	147	146	139	120	109	134	134	210	205	203
Epoxiconazole	22	31	47	46	41	32	31	74	60	56	67	65
Ethofumesate	60	166	561	556	487	304	255	351	596	920	809	833
Fenpropimorph	<10	<5	13	13	<5	11	9	<5	<5	13	17	14
Florasulam	<100	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Fluazifop (free acid)	<40	28	170	183	135	87	58	72	276	369	332	365
Fluazinam	<200	58	58	42	20	41	37	148	158	110	122	148
Flufenacet	<8	185	167	166	119	101	83	619	276	255	248	302
Fluopicolide	<8	575	980	860	738	457	368	1747	2107	1896	1657	1910
Flupyrsulfuron-methyl	<10	<9	<10	<9	<9	<9	<9	<9	<9	<9	<9	<9
Foramsulfuron	<9	15	<9	<9	<9	<9	<9	36	<9	15	<9	14
Iodosulfuron-methyl	<10	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15
Isoproturon	<6	<4	<6	<4	<4	<4	<4	<4	<4	<4	<4	<4
Lenacil	<10	235	814	890	705	468	359	421	898	1280	1278	1402
Mandipropamid	<8	13	30	29	19	11	12	95	162	157	131	114
Mecoprop	<100	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40
Mesosulfuron-methyl	<9	<8	<9	<8	<8	<8	<8	<8	<8	<8	<8	<8
Metamitron	61	108	214	216	199	182	166	217	204	217	238	270
Metolachlor	<8	13	18	18	18	16	13	35	38	25	21	29
Metrafenone	<15	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Metribuzin	159	162	360	334	286	252	216	461	652	550	514	574
Napropamide	<8	12	12	14	12	13	12	13	11	12	<10	12
Nicosulfuron	<10	18	130	145	98	111	86	127	68	101	130	135
Pencycuron	283	142	910	853	791	572	490	474	1543	1502	1569	1590
Propamocarb	<50	2496	19800	12205	4323	2731	1966	20630	35020	25631	23553	23474
Propiconazole	<15	<8	12	13	11	11	9	<8	16	18	17	18
Prosulfocarb	241	112	479	11	377	287	252	308	541	639	644	536
Prothioconazole	<100	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Pyraclostrobin	<50	<9	<15	<9	<9	<9	<9	<9	<9	<9	<9	<9
Spiroxamine	<100	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25
Tembotrione	95	<8	33	35	22	23	14	47	23	18	32	35
Terbuthylazine	10362	632	628	558	450	402	340	2331	1751	1524	1345	1336
Thiacloprid	<15	7	<15	<5	<5	<5	<5	8	5	<5	<5	<5
Thiamethoxam	<10	<8	<10	23	25	23	<8	<8	<8	14	19	26
Trifloxystrobin	<50	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
CT-TP-R417888	<45	1484	1587	1869	<55	1431	1455	1654	2360	2382	2119	2104
CT-TP-R471811	<100	3777	1619	2075	1184	2946	3335	1304	646	957	1000	961
CT-TP-R611968	<15	31	35	34	22	34	43	31	35	43	39	41
CT-TP-SYN507900	<50	253	221	288	110	289	322	98	72	138	154	152
Metamitron-desamino	25	542	2369	2568	2409	1731	1169	994	3424	4140	4094	4006
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	31	54	36	32	37

Event	12	12	12	12	12	12	12	12	12	12	12	12	12
Sampling site	**ST**	**ST**	**ST**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**
Sample type	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI	TI
Sampling end	15.06 23:33	16.06 00:03	16.06 00:33	15.06 18:33	15.06 23:03	16.06 00:03	16.06 01:03	15.06 19:03	15.06 19:33	15.06 20:03	15.06 20:33	15.06 21:03	15.06 21:33
Sampling start	15.06 23:03	15.06 23:33	16.06 00:03	15.06 18:03	15.06 22:33	15.06 23:33	16.06 00:33	15.06 18:33	15.06 19:03	15.06 19:33	15.06 20:03	15.06 20:33	15.06 21:03
Measurement set	2	2	2	2	2	2	2	2	2	2	2	2	2
Azoxystrobin	13	13	12	10	<9	<9	<9	12	12	19	13	10	<9
Bixafen	44	48	44	69	<20	<20	<20	40	40	47	49	34	<20
Boscalid	147	127	126	19	<10	<10	<10	15	18	15	20	18	14
Carfentrazone-ethyl	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20
Chlortoluron	253	237	217	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
Cymoxanil	25	33	22	51	87	51	48	129	282	247	382	340	335
Cyproconazole	23	27	23	13	<8	<8	<8	21	19	24	15	<8	<8
Difenoconazole	269	230	204	16	10	<8	<8	23	30	68	60	42	23
Diflufenican	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Dimethachlor	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Dimethenamid	166	161	176	312	87	83	90	561	357	509	255	138	87
Epoxiconazole	70	68	62	115	12	9	10	60	58	54	46	27	16
Ethofumesate	824	757	702	108	51	50	39	97	67	66	76	79	75
Fenpropimorph	17	11	14	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Florasulam	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Fluazifop (free acid)	250	283	227	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Fluazinam	95	86	80	<8	10	<8	<8	<8	<8	<8	<8	9	15
Flufenacet	275	256	214	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20
Fluopicolide	1642	1437	1386	19	11	<9	<9	24	30	25	23	27	26
Flupyrsulfuron-methyl	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Foramsulfuron	18	<9	10	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Iodosulfuron-methyl	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15
Isoproturon	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4
Lenacil	1183	1077	1057	21	28	34	44	<15	<15	<15	22	<15	<15
Mandipropamid	94	91	64	16	13	12	11	43	68	140	132	58	38
Mecoprop	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40
Mesosulfuron-methyl	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Metamitron	251	223	229	150	67	60	63	134	143	93	116	96	82
Metolachlor	21	21	20	25	<8	<8	<8	25	21	14	12	11	<8
Metrafenone	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Metribuzin	522	532	461	144	1889	2290	2353	230	433	596	871	1172	1188
Napropamide	12	12	12	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Nicosulfuron	142	133	146	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Pencycuron	1444	1220	1114	302	344	339	323	809	490	444	604	593	398
Propamocarb	28580	21976	19250	134	58	44	39	211	284	246	159	152	137
Propiconazole	17	14	16	9	<8	<8	<8	<8	18	<8	<8	<8	<8
Prosulfocarb	627	511	527	24	<8	<8	<8	26	27	22	21	12	11
Prothioconazole	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Pyraclostrobin	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Spiroxamine	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25
Tembotrione	25	28	32	67	<8	<8	<8	117	41	78	27	<8	<8
Terbuthylazine	1064	1060	892	1742	88	57	40	3332	2626	4616	1947	664	312
Thiacloprid	<5	7	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Thiamethoxam	24	18	28	<8	43	39	28	<8	15	17	35	58	48
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
CT-TP-R417888	1935	2079	2027	697	1686	1626	1627	371	324	328	636	1092	1250
CT-TP-R471811	1083	1195	1439	2127	4124	4395	4465	1241	750	726	1376	2271	3020
CT-TP-R611968	37	37	38	26	79	63	62	24	31	33	57	91	88
CT-TP-SYN507900	161	166	209	162	494	372	385	106	86	88	191	359	391
Metamitron-desamino	3473	3133	2830	136	60	75	72	106	88	65	66	69	66
Metolachlor-OXA	20	19	16	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	12	12	12	12	12	12	13	13	13	13	13	13	13	13
Sampling site	**CS**	**CS**	**I1**	**I2**	**I3**	**I4**	**ST**	**ST**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**
Sample type	TI	TI	WL	WL	GR	WL	TI	TI	TI	TI	TI	TI	TI	TI
Sampling end	15.06 22:03	15.06 22:33	NA	NA	NA	NA	02.07 00:00	02.07 16:00	01.07 19:17	01.07 19:54	01.07 20:24	01.07 20:54	01.07 21:21	02.07 00:56
Sampling start	15.06 21:33	15.06 22:03	15.06 18:01	15.06 18:01	17.06 11:05	15.06 18:01	01.07 16:00	02.07 08:00	01.07 18:47	01.07 19:17	01.07 19:54	01.07 20:24	01.07 20:54	02.07 00:26
Measurement set	2	2	1	1	1	1&3	3	3	3	3	3	3	3	3
Azoxystrobin	<9	<9	14	<7	<7	76	69	565	<6	<6	<6	<6	<6	<6
Bixafen	<20	<20	284	<8	251	68	7	14	29	45	42	18	<5	31
Boscalid	<10	<10	<10	<10	<10	<10	18	49	<4	5	4	<4	<4	9
Carfentrazone-ethyl	<20	<20	<10	<10	<10	<15	<15	<15	<15	<15	<15	<15	<15	<15
Chlortoluron	<6	<6	<10	<10	<10	<10	<6	<6	<6	<6	<6	<6	<6	<6
Cymoxanil	117	146	<9	<9	<9	156	<9	<9	10	<9	<9	<9	<9	<9
Cyproconazole	<8	<8	<8	<8	<8	316	85	537	<4	<4	<4	<4	<4	<4
Difenoconazole	19	14	<25	<25	<25	<25	<8	60	<8	<8	<8	<8	<8	9
Diflufenican	<200	<200	<500	<500	<500	<500	<100	<100	<100	<100	<100	<100	<100	<100
Dimethachlor	<9	<9	<6	<6	<6	<10	<10	<10	<10	<10	<10	<10	<10	<10
Dimethenamid	74	79	<9	<9	<9	7698	<4	95	58	93	42	20	10	5675
Epoxiconazole	12	12	390	<7	17	20	9	25	12	34	26	13	7	44
Ethofumesate	57	60	106	31	33	65	114	219	78	1483	696	283	106	1794
Fenpropimorph	<5	<5	<6	<6	<6	<10	<10	<10	<10	<10	<10	<10	<10	<10
Florasulam	<50	<50	<100	<100	<100	<100	<20	<20	<20	<20	<20	<20	<20	<20
Fluazifop (free acid)	<9	<9	<40	<40	<40	<40	<10	<10	<10	<10	<10	<10	<10	<10
Fluazinam	26	16	<200	<200	<200	<200	14	18	<4	<4	<4	<4	<4	<4
Flufenacet	<20	<20	<8	<8	<8	<8	63	379	<6	<6	<6	<6	<6	<6
Fluopicolide	22	22	38	27	31	49	45	100	<2	<2	<2	<2	<2	<2
Flupyrsulfuron-methyl	<9	<9	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Foramsulfuron	<9	<9	<9	<9	<9	<9	10	13	<4	<4	<4	<4	<4	<4
Iodosulfuron-methyl	<15	<15	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Isoproturon	<4	<4	<2	<2	<2	<6	<6	<6	<6	<6	<6	<6	<6	<6
Lenacil	21	23	<10	<10	<10	<10	64	64	<8	<8	<8	<8	<8	9
Mandipropamid	25	17	<8	<8	<8	<8	24	167	<4	<4	<4	<4	<4	5
Mecoprop	<40	<40	<100	<100	<100	<100	<5	74	<5	<5	<5	<5	<5	<5
Mesosulfuron-methyl	<8	<8	<8	<8	<8	<9	<9	<9	<9	<9	<9	<9	<9	<9
Metamitron	75	69	64	<20	<20	99	37	222	108	2410	1026	391	172	2703
Metolachlor	<8	<8	24	34	23	33	<4	14	20	22	14	8	<4	16
Metrafenone	<9	<9	90	<15	<15	<15	<6	<6	<6	<6	<6	<6	<6	<6
Metribuzin	1207	1639	<9	<9	<9	31	23	46	23	7	6	5	5	10
Napropamide	<10	<10	<8	<8	<8	<8	<6	6	<6	<6	<6	<6	<6	<6
Nicosulfuron	<8	<8	<10	<10	<10	<10	<6	66	17	26	9	<6	<6	703
Pencycuron	328	360	<15	<15	<15	67	34	67	224	175	110	113	66	220
Propamocarb	144	105	<5	<5	<5	376	271	604	<50	<50	<50	<50	<50	<50
Propiconazole	<8	<8	<15	<15	<15	<15	<6	<6	<6	<6	<6	<6	<6	<6
Prosulfocarb	10	9	<15	<15	<15	48	20	54	3	2	3	<2	<2	5
Prothioconazole	<200	<200	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Pyraclostrobin	<9	<9	<50	<50	<50	<50	<15	<15	<15	<15	<15	<15	<15	<15
Spiroxamine	<25	<25	<100	<100	<100	<100	<20	<20	<20	<20	<20	<20	<20	<20
Tembotrione	<8	<8	<50	<50	<50	3977	13	73	<8	<8	<8	<8	<8	335
Terbuthylazine	176	114	58	31	56	61730	143	980	195	334	163	70	39	7871
Thiacloprid	<5	<5	<7	<7	<7	<15	28	<15	<15	<15	<15	<15	<15	<15
Thiamethoxam	45	33	<10	<10	<10	<10	35	14	<10	<10	<10	<10	<10	<10
Trifloxystrobin	<10	<10	<50	<50	<50	<50	<10	124	<10	<10	<10	<10	<10	<10
CT-TP-R417888	1317	1540	813	<45	465	<45	1574	1223	988	544	881	1199	1410	522
CT-TP-R471811	3328	3975	<100	<100	<100	<100	3372	2220	2865	1586	2196	2992	3465	1307
CT-TP-R611968	68	70	<15	<15	<15	<15	16	<5	13	<5	<5	14	11	7
CT-TP-SYN507900	412	416	<50	<50	<50	<50	121	119	243	129	169	238	300	124
Metamitron-desamino	62	62	72	<10	<10	55	125	252	40	491	274	142	51	651
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	13	13	13	13	13	13	13	13	13	13	13	14	14	14
Sampling site	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**CS**	**I1**	**I2**	**I3**	**I4**	**ST**	**ST**	**ST**
Sample type	TI	TI	TI	TI	TI	TI	TI	WL	WL	WL	WL	TI	TI	TI
Sampling end	02.07 01:26	02.07 01:56	02.07 02:26	02.07 03:26	02.07 04:26	02.07 06:56	02.07 07:26	NA	NA	NA	NA	06.07 16:00	07.07 00:00	07.07 08:00
Sampling start	02.07 00:56	02.07 01:26	02.07 01:56	02.07 02:26	02.07 03:26	02.07 04:26	02.07 06:56	01.07 19:25	02.07 11:35	02.07 10:50	02.07 10:20	06.07 08:00	06.07 16:00	07.07 00:00
Measurement set	3	3	3	3	3	3	3	1&3	2	2	2&3	3	3	3
Azoxystrobin	<6	<6	<6	<6	<6	<6	<6	<7	<9	13	72	30	168	87
Bixafen	21	14	7	<5	<5	<5	<5	92	<20	333	74	8	13	13
Boscalid	7	<4	<4	<4	<4	<4	<4	<10	<10	<10	<10	13	178	89
Carfentrazone-ethyl	<15	<15	<15	<15	<15	<15	<15	<15	<20	<20	<20	<15	<15	<15
Chlortoluron	<6	<6	<6	<6	<6	<6	<6	<10	<6	<6	<6	14	22	109
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	30	<9	<9	<9
Cyproconazole	<4	<4	<4	<4	<4	<4	<4	<8	<8	11	192	45	301	168
Difenoconazole	<8	<8	<8	<8	<8	<8	<8	<25	<8	28	137	<8	24	18
Diflufenican	<100	<100	<100	<100	<100	<100	<100	<500	<200	<200	<200	<100	<100	<100
Dimethachlor	<10	<10	<10	<10	<10	<10	<10	<10	<9	<9	<10	<10	<10	<10
Dimethenamid	2532	415	112	54	34	27	24	18	11	10	3452	<4	4	<4
Epoxiconazole	33	14	<6	<6	<6	<6	<6	598	<8	12	23	18	29	27
Ethofumesate	844	389	105	45	28	26	18	5920	31	25	61	106	192	284
Fenpropimorph	<10	<10	<10	<10	<10	<10	<10	<10	<5	<5	<10	<10	<10	<10
Florasulam	<20	<20	<20	<20	<20	<20	<20	<100	<50	<50	<50	<20	<20	<20
Fluazifop (free acid)	<10	<10	<10	<10	<10	<10	<10	<40	<9	<9	<10	<10	38	117
Fluazinam	<4	<4	<4	<4	<4	<4	<4	<200	<8	<8	<8	<4	11	<4
Flufenacet	<6	<6	<6	<6	<6	<6	<6	<8	<20	<20	<20	17	224	117
Fluopicolide	<2	<2	<2	<2	<2	<2	<2	30	54	84	103	32	92	66
Flupyrsulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<9	<9	<10	<10	<10	<10
Foramsulfuron	<4	<4	<4	<4	<4	<4	<4	<9	<9	<9	<9	<4	28	11
Iodosulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<15	<15	<15	<10	<10	<10
Isoproturon	<6	<6	<6	<6	<6	<6	<6	<6	<4	<4	<6	<6	<6	<6
Lenacil	<8	<8	<8	<8	<8	<8	<8	<10	<15	<15	<15	90	173	431
Mandipropamid	6	<4	<4	<4	<4	<4	<4	<8	<9	<9	192	4	331	121
Mecoprop	<5	<5	<5	<5	<5	<5	<5	<100	<40	<40	<40	<5	52	22
Mesosulfuron-methyl	<9	<9	<9	<9	<9	<9	<9	<9	<8	<8	<9	<9	<9	<9
Metamitron	1449	721	158	70	37	30	31	7889	11	11	45	25	96	151
Metolachlor	16	10	4	<4	<4	<4	<4	37	26	14	17	<4	<4	6
Metrafenone	<6	<6	<6	<6	<6	<6	<6	<15	<9	<9	<9	<6	<6	<6
Metribuzin	16	17	17	17	12	9	6	<9	<7	<7	63	16	35	24
Napropamide	<6	<6	<6	<6	<6	<6	<6	24	<10	<10	<10	<6	11	7
Nicosulfuron	296	53	16	7	<6	<6	<6	<10	<8	<8	<8	<6	21	17
Pencycuron	557	454	358	280	250	228	236	<15	<9	13	191	52	84	136
Propamocarb	<50	<50	<50	<50	<50	<50	<50	<50	48	37	750	<50	265	124
Propiconazole	<6	<6	<6	<6	<6	<6	<6	<15	<8	<8	<8	<6	<6	<6
Prosulfocarb	6	<2	<2	<2	<2	<2	<2	<15	20	19	344	14	39	52
Prothioconazole	<100	<100	<100	<100	<100	<100	<100	<100	<200	<200	<200	<100	<100	<100
Pyraclostrobin	<15	<15	<15	<15	<15	<15	<15	<50	<9	<9	<15	<15	<15	<15
Spiroxamine	<20	<20	<20	<20	<20	<20	<20	<100	<25	<25	<25	<20	<20	<20
Tembotrione	150	27	<8	<8	<8	<8	<8	<50	<8	<8	515	<8	22	11
Terbuthylazine	3840	634	209	98	59	45	43	100	64	70	31075	91	451	310
Thiacloprid	<15	<15	<15	<15	<15	<15	<15	<15	<5	<5	105	<15	<15	<15
Thiamethoxam	<10	<10	<10	<10	<10	<10	<10	<10	<8	<8	<10	49	69	89
Trifloxystrobin	<10	<10	<10	<10	<10	<10	<10	<50	<10	<10	<10	<10	12	<10
CT-TP-R417888	732	1111	1399	1553	1583	1538	1569	282	<55	390	<55	1713	1519	1059
CT-TP-R471811	1880	2727	1984	3690	3716	2808	3870	224	<15	<15	<100	4072	2659	2112
CT-TP-R611968	<5	<5	<5	<5	<5	<5	<5	<15	<10	37	12	<5	32	<5
CT-TP-SYN507900	161	265	269	317	344	318	346	<50	<20	<20	<20	224	157	159
Metamitron-desamino	425	186	58	26	10	6	7	4912	30	17	85	346	732	2161
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	14	14	14	14	14	15	15	15	15	16	16	16	16	17
Sampling site	**ST**	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**I3**	**I4**	**I2**	**I1**	**I3**	**I4**	**I1**
Sample type	TI	WL	GR	GR	GR	WL	WL	GR	GR	GR	GR	GR	GR	WL
Sampling end	07.07 16:00	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Sampling start	07.07 08:00	06.07 14:23	10.07 13:00	10.07 13:15	10.07 12:00	14.07 22:40	14.07 22:40	16.07 11:15	16.07 12:05	29.07 09:22	29.07 08:20	29.07 09:00	29.07 08:41	06.08 09:21
Measurement set	3	1	2	2	2&3	1	1	1	1	2	2	1	1	1
Azoxystrobin	138	<7	<9	16	75	<7	<7	20	33	<9	<9	25	39	13
Bixafen	22	75	<20	358	72	50	<8	323	63	<20	40	397	66	<8
Boscalid	124	17	<10	<10	<10	<10	<10	<10	<10	<10	102	<10	<10	66
Carfentrazone-ethyl	<15	<10	<20	<20	<20	<10	<10	<10	<10	<20	<20	<10	<10	<10
Chlortoluron	185	<10	<6	13	<6	<10	<10	17	<10	<6	<6	25	<10	<10
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	285	<8	<8	<8	182	<8	<8	<8	93	38	<8	12	109	<8
Difenoconazole	23	<25	<8	38	71	55	<25	54	146	<8	14	55	74	1327
Diflufenican	<100	<500	<200	<200	<200	<500	<500	<500	<500	<200	<200	<500	<500	<500
Dimethachlor	<10	<6	<9	<9	<10	<6	<6	<6	<6	<9	<9	<6	<6	<6
Dimethenamid	<4	<9	10	<8	2668	<9	<9	<9	248	<8	<8	<9	193	10
Epoxiconazole	34	1471	10	12	22	523	21	<7	24	13	413	<7	25	380
Ethofumesate	357	3649	39	29	67	1923	<9	<9	51	22	1074	<9	68	1080
Fenpropimorph	<10	<6	24	<5	<10	<6	<6	<6	<6	17	<5	<6	<6	<6
Florasulam	<20	<100	<50	<50	<50	<100	<100	<100	<100	<50	<50	<100	<100	<100
Fluazifop (free acid)	117	<40	<9	<9	<10	<40	<40	<40	<40	<9	<9	<40	<40	<40
Fluazinam	<4	<200	<8	<8	<8	<200	<200	<200	<200	<8	<8	<200	<200	<200
Flufenacet	192	<8	<20	<20	<20	<8	<8	<8	<8	<20	<20	<8	<8	<8
Fluopicolide	110	26	71	135	161	<8	56	140	124	59	14	193	124	27
Flupyrsulfuron-methyl	<10	<10	<9	<9	<10	<10	<10	<10	<10	<9	<9	<10	<10	<10
Foramsulfuron	68	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Iodosulfuron-methyl	<10	<10	<15	<15	<15	<10	<10	<10	<10	<15	<15	<10	<10	<10
Isoproturon	<6	4	<4	<4	<6	<2	<2	<2	<2	<4	<4	<2	<2	2
Lenacil	715	<10	<15	<15	27	<10	<10	<10	<10	<15	<15	<10	<10	<10
Mandipropamid	162	<8	<9	<9	66	59	<8	<8	167	<9	<9	<8	<8	41
Mecoprop	21	<100	<40	49	<40	<100	<100	<100	<100	<40	<40	<100	<100	<100
Mesosulfuron-methyl	9	<8	<8	<8	<9	<8	<8	<8	<8	<8	<8	<8	<8	<8
Metamitron	115	2190	<7	<7	30	740	<20	<20	22	<7	339	<20	<20	170
Metolachlor	<4	<8	25	12	18	<8	38	<8	<8	12	11	<8	<8	<8
Metrafenone	<6	<15	<9	<9	<9	<15	<15	<15	<15	<9	<9	<15	<15	<15
Metribuzin	39	<9	<7	<7	66	<9	<9	<9	31	<7	<7	<9	34	<9
Napropamide	12	15	<10	<10	<10	<8	<8	<8	<8	<10	16	<8	<8	15
Nicosulfuron	<6	<10	<8	<8	<8	<10	<10	<10	<10	<8	<8	<10	<10	<10
Pencycuron	171	<15	<9	<9	182	<15	<15	<15	124	<9	<9	<15	121	<15
Propamocarb	145	<5	66	46	719	<5	<5	<5	274	35	<25	<5	175	<5
Propiconazole	<6	<15	<8	<8	<8	<15	<15	<15	<15	<8	10	<15	<15	<15
Prosulfocarb	64	<15	23	26	474	<15	<15	<15	128	16	<8	<15	192	<15
Prothioconazole	<100	<100	<200	<200	<200	<100	<100	<100	<100	<200	<200	<100	<100	<100
Pyraclostrobin	<15	<50	<9	<9	<15	<50	<50	<50	<50	<9	<9	<50	<50	<50
Spiroxamine	<20	<100	<25	<25	<25	<100	<100	<100	<100	<25	<25	<100	<100	<100
Tembotrione	19	<50	<8	<8	456	<50	<50	<50	82	<8	<8	<50	102	<50
Terbuthylazine	449	52	69	66	26131	29	24	47	4136	21	14	41	4313	48
Thiacloprid	17	<7	<5	<5	81	<7	<7	<7	117	<5	<5	<7	84	<7
Thiamethoxam	60	<10	<8	<8	<10	<10	<10	<10	<10	<8	<8	<10	<10	<10
Trifloxystrobin	<10	<50	<10	<10	<10	<50	<50	<50	<50	<10	<10	<50	<50	<50
CT-TP-R417888	1497	103	<55	374	<55	108	<45	444	82	<55	59	389	<45	<45
CT-TP-R471811	2821	<100	<15	<15	<100	<100	<100	<100	<100	<15	84	<100	<100	151
CT-TP-R611968	<5	<15	<10	35	13	<15	<15	<15	<15	<10	12	<15	<15	<15
CT-TP-SYN507900	146	<50	<20	<20	<20	<50	<50	<50	<50	<20	<20	<50	<50	<50
Metamitron-desamino	2837	2118	51	30	123	1056	21	19	26	58	585	31	69	280
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Event	17	17	17	18	18	18	18	19	19	19	19
Sampling site	**I2**	**I3**	**I4**	**I1**	**I2**	**I3**	**I4**	**I1**	**I2**	**I3**	**I4**
Sample type	WL	GR	GR	WL	WL	WL	WL	GR	GR	GR	GR
Sampling end	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Sampling start	06.08 09:21	08.08 09:25	08.08 09:15	10.08 09:29	10.08 09:29	10.08 09:29	10.08 09:29	20.08 09:10	20.08 09:49	20.08 09:35	20.08 09:25
Measurement set	1	1	1	1	1	1	1	1	1	1	1
Azoxystrobin	<7	25	24	<7	<7	<7	19	<7	<7	15	18
Bixafen	<8	227	50	<8	<8	149	60	<8	<8	126	47
Boscalid	38	<10	<10	22	30	<10	<10	18	16	<10	<10
Carfentrazone-ethyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Chlortoluron	<10	27	<10	<10	<10	<10	<10	<10	<10	<10	<10
Cymoxanil	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Cyproconazole	112	<8	49	<8	81	<8	35	<8	38	<8	39
Difenoconazole	<25	56	60	270	47	<25	59	140	<25	<25	51
Diflufenican	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500	<500
Dimethachlor	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
Dimethenamid	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Epoxiconazole	16	15	20	112	<7	<7	16	90	13	21	36
Ethofumesate	<9	<9	<9	296	<9	<9	<9	175	<9	<9	<9
Fenpropimorph	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	91
Florasulam	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Fluazifop (free acid)	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40	<40
Fluazinam	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Flufenacet	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Fluopicolide	58	211	45	<8	39	38	30	<8	20	35	23
Flupyrsulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Foramsulfuron	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Iodosulfuron-methyl	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Isoproturon	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Lenacil	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Mandipropamid	35	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Mecoprop	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Mesosulfuron-methyl	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Metamitron	<20	<20	<20	69	<20	<20	<20	40	<20	<20	<20
Metolachlor	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Metrafenone	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15
Metribuzin	<9	<9	12	<9	<9	<9	<9	<9	<9	<9	<9
Napropamide	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Nicosulfuron	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Pencycuron	<15	<15	64	<15	<15	<15	54	<15	<15	<15	47
Propamocarb	<5	<5	37	<5	<5	<5	24	<5	<5	<5	24
Propiconazole	<15	<15	<15	<15	42	<15	<15	<15	<15	<15	<15
Prosulfocarb	<15	41	52	<15	<15	<15	<15	<15	<15	<15	<15
Prothioconazole	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Pyraclostrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Spiroxamine	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Tembotrione	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Terbuthylazine	17	37	188	<9	<9	<9	66	<9	<9	<9	47
Thiacloprid	<7	<7	11	<7	<7	<7	<7	<7	<7	<7	<7
Thiamethoxam	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Trifloxystrobin	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
CT-TP-R417888	<45	319	<45	<45	<45	<45	<45	<45	<45	<45	<45
CT-TP-R471811	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
CT-TP-R611968	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15	<15
CT-TP-SYN507900	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Metamitron-desamino	20	34	11	61	<10	<10	<10	38	<10	<10	<10
Metolachlor-OXA	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Curriculum vitae

Full name: Urs Thomas Schönenberger

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Education and work experience

2016 – 2021 **PhD candidate, Environmental Engineering**
ETH Zürich and Department of Environmental Chemistry, Swiss Federal Institute of Aquatic Science and Technology (Eawag), Switzerland

2015 – 2016 **Research associate**
Department of Environmental Chemistry, Swiss Federal Institute of Aquatic Science and Technology (Eawag), Switzerland

2013 – 2015 **Master of Science in Environmental Engineering**
ETH Zürich, Switzerland

2012 – 2013 **Civil service**
Water division, Federal Office for the Environment, Bern

2009 – 2012 **Bachelor of Science in Environmental Engineering**
ETH Zürich, Switzerland

2005 – 2009 **High school**
Kantonsschule am Burggraben, St. Gallen, Switzerland

Publications

Schönenberger, U.; Beck, B.; Dax, A.; Vogler, B.; Stamm, C. (2022): Pesticide concentrations in agricultural storm drainage inlets of a small Swiss catchment, *Environ. Sci. Pollut. Res.*, DOI: 10.1007/s11356-022-18933-5

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Munz, N. A.; Burdon, F. J.; de Zwart, D.; Junghans, M.; Melo, L.; Reyes, M.; Schönenberger, U.; Singer, H. P.; Spycher, B.; Hollender, J.; Stamm, C. (2017): Pesticides drive risk of micropollutants in wastewater-impacted streams during low flow conditions, *Water Research*, 110, 366-377, DOI: 10.1016/j.watres.2016.11.001

Extra task for attentive readers

As a child, my brother painted this landscape on a potholder as a christmas present for our parents. If you look closely, you can spot his initials “M.S.” on the bottom right of the potholder. The drawing shows a typical rural landscape in Switzerland, reduced to those landscape elements that seemed important to my brother.

Amazingly, in his simplified landscape drawing, he only used elements that are also important in the context of pesticide transport to surface waters via hydraulic shortcuts: A river with fishes (an aquatic ecosystem), fields with flowers (agricultural crops), hedges (that may intercept spray drift), and an asphalt road (where most hydraulic shortcuts are located).

If you have read this thesis, take this drawing with you to your next coffee break. Using the drawing, explain to the first person you meet what hydraulic shortcuts are and why they should not be overlooked.