Emissions during hot-recycling of recycled asphalt pavement containing tar

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ABSTRACT: Hot recycling in pavement construction leads to the emission of hazardous compounds when tar-containing recycled asphalt pavement (RAP) is used. This is due to the relatively high content of substances such as phenols and polycyclic aromatic hydrocarbons (PAH) in tar. The latter are of special interest, because some PAHs have carcinogenic and/or mutagenic properties. High tar content in combination with high pavement temperatures during construction increases the potential health risk for workmen. Switzerland does not currently consider a complete ban of RAP in hot recycling for economical and ecological reasons. Since RAP might always contain some tar, there is a need to fix a scientifically sound limit for the maximum tar content. However, scientific data on the emissions of PAH, as well as the resulting occupational health risk were not sufficient. Thus, the present research project was carried out to close this gap.

Different test parameters such as temperature, PAH content and viscosity were varied in the laboratory test program to investigate their influence on the resulting emissions. It was shown that temperature is the major factor determining the emissions in the 140 to 240°C range. A PAH content of up to 20'000 ppm EPA-PAH in the RAP had no significant effect on total particulate matter (TPM) emissions. However, emissions of PAH in the fumes increased proportionally with the PAH concentration in the RAP.

Three field tests were carried out with RAP containing up to 5600 ppm EPA-PAH. Occupational health data for the workmen were collected with personal samplers. Concentrations of TPM were with 2-4 mg/m^3 close to the occupational health limit value for bitumen fumes (10 mg/m^3). Concentrations of Benzo(a)pyrene (BaP), which is often used as a lead compound, were in all cases below the occupational health limit value of 2 $\mu g/m^3$ by at least a factor of three.

This study indicates that the current Swiss recommended maximum value of 5000 ppm for EPA-PAH in the binder of RAP may be considered as adequate. However, conditions in the field trials did not include a worst-case scenario and the gap to certain occupational health limit values is small.

1. Introduction

Switzerland has a long tradition of asphalt recycling and introduced in 1993 standards on recycling materials including recycled asphalt pavement (RAP) [1]. These standards describe the application field of recycled materials and defined some requirements on their purity. People were convinced that only material with a declared and monitored quality could achieve a good acceptance of recycled products and get rid of the image of waste material. Recent past has shown that this was the right approach, because recycled products are widely accepted and used throughout Switzerland.

Already the first version of the standard on RAP treated some aspects of possible environmental impacts coming from tar-containing material. The carcinogenic and mutagenic potential of some polycyclic aromatic hydrocarbons (PAH), Benzo(a)pyrene in particular, became public. In contrast to bitumen, tar contains large amounts of PAH amounting up to 20 mass-%. Due to the good mechanical properties of tar, tar-containing binders were used in large quantities in some parts of Switzerland until 1990. For this reason, tar is present in RAP in varying quantities.

At the time the recycling standards were generated, analytical chemical methods for the quantitative analysis of the toxic PAH were tedious and expensive. Therefore RAP was divided into three classes: RAP without tar, RAP with small amounts of tar and RAP with large amounts of tar. On first sight this makes sense, as originally binders were categorised by Swiss standards into tar, bitumen-tar, tar-bitumen and bitumen with defined tar contents. However, there were several reasons why this concept did not work properly:

1. Road layers are typically milled without considering the individual pavement layers as, in general, no information about the type of binders used in the different layers is available.
2. In recycling plants, low and high-contaminated RAP are stored together on one heap.
3. Nowadays, some pavements have already been recycled for the second or even the third time. Because RAP was originally added only in proportions of about 10 - 25 %, tar in the recycled layers was gradually diluted, resulting in pavement material with a tar content that is no longer consistent with the original classification.
4. During the lifetime of a road, trenches are excavated to repair water pipes, new cables installed for television networks, etc. Especially in cities, road pavements look like a patchwork after a short time.

To overcome this problem of non-measurable limits, the Swiss Federal Office for the Environment (FOEN) enacted a guideline on the re-use of waste and fixed a limit value of 5000 ppm EPA-PAHs in the binder of new pavements [2]. However, this limit value was not based on occupational health factors but was a compromise between political, economical and ecological considerations. No wonder then, that different interests have challenged this value to assign a lower or higher limit value.

This was the reason why Empa launched a research project to provide the scientific knowledge to fix a limit value based on occupational health measurements.

The research project was divided into two parts: A laboratory study to investigate the effect and importance of various parameters, such as PAH concentration, temperature and viscosity, on the emission of fumes and PAHs. Field tests were then carried out to get information on the emission behaviour in the field and to obtain personal occupation data.

2. Laboratory testing

2.1 Fume generation

The first challenge was the production of fumes in the laboratory equivalent to the fumes during paving. The correct fume production procedure in the laboratory has previously been heavily disputed [3] as a result of published emission data for pure bitumen [4, 5]. For these studies, bitumen was heated to very high temperatures to get enough condensates out of the emitted fumes for skin tests on mice. The higher the temperature, the more fumes are produced, but also the more cracking is taking place, producing toxic decomposition products [6]. This results in fume compositions radically different from bitumen fumes during paving.

For this reason it is essential to use a fume generator producing fumes in compositions that match the fumes emitted during paving as closely as possible. Still, no standard fume composition can be defined as the fumes are dependant on the type of bitumen used, as well as on temperature, paving procedures and other factors.

A literature search revealed two predominant fume generation methods which have been validated in the field. A simple fume generator was designed and validated by Brandt et. al.. It consists of an open glass flask containing the bitumen, which is heated in a temperature controlled heating mantle [7-9]. Fumes are collected by two or more sampling devices mounted on top of the flask. Another fume generator was developed by Kurek et al. to produce fumes for animal inhalation studies to test the carcinogenic potential of bitumen fumes [10, 11]. This fume generator consists of a hot inclined surface enclosed in a sealed box. Bitumen flows in a thin film down the plane under rigidly controlled conditions [12, 13]. Apart from the very sensitive test conditions, a large amount of bitumen is needed. But fumes produced by this installation are constant in composition throughout the whole experiment, which is not the case with the simple fume generator of Brandt, where the fume composition changes over time, as the higher volatile components are depleted. Despite this, the fume generator described by Brandt was selected and adapted to meet the requirements of our laboratory study (Figure 1), as in our case, the fume was not detected continuously but sampled in batches. Another reason for this choice was the scarcity of tar binders, as they are no longer produced in Switzerland.

2.2 Sampling and chemical analysis

Figure 1: Fume generator without cardboard for the laboratory tests

Despite the simple construction of the fume generator, sampling of the emitted fumes proved difficult. The slightest air movement caused an inhomogeneous fume repartition above the vessel. To overcome this problem, a small, dedicated fan was mounted vertically above the fume generator and the fume generator enclosed with cardboard rising 15 cm above the vessel. Homogeneity was confirmed through repeated measurements from two sets of sampling units set up diagonally over the flask opening.

Fumes of tar-containing binders contain both high- (2- and 3-ring) and low-volatile (4- to 6-ring) PAHs. This required a sampling unit consisting of two parts: a teflon-coated glass filter to collect the particle-bound and condensed PAHs, and a backup adsorption tube (ORBO-43, XAD-2) placed between the filter and the sampling pump for volatile PAHs.

Filter residues were weighed for the gravimetric determination of total particulate matter (TPM). Afterwards, filters and adsorption tubes were extracted with toluene and the combined solution was analysed by GC/MS (Gas chromatography combined with mass spectrometry) and RP-HPLC (Reversed phase high performance liquid chromatography). Concentrations of all 16 EPA-PAHs in the fume were determined both in the laboratory and in the field tests. The sampling and chemical analysis are described in more detail in [14] and in the original report [15].

2.3 Test program

In a first set of experiments, pure tar hv54 and a pure bitumen were blended to study mixtures with different PAH contents. The influence of temperature on the emission behaviour was studied between 140 and 240 °C. This is the range of paving temperatures used in the field for hot mixes and mastic asphalt. However, at temperatures above 210°C, fume generation was so strong, that the described sampling units were overloaded. To study emissions from mastic asphalt in detail, the sampling procedure and the fume generation would have to be adapted.

Table 1: Test parameters of the laboratory experiments

Binder composition (% w/w)			EPA-PAH content of the binder (mg/kg)	Test Temperature (°C)
Tar hv54	Tar binder TB 2000	Bitumen 50/70		
0	33	67	5100	140
0	33	67	5100	160
0	33	67	5100	180
0	33	67	5100	210
0	0	100	105	160
1	0	99	2430	160
2.5	0	97.5	5930	160
5	0	95	11'800	160
10	0	90	23'400	160
20	0	80	46'700	160
50	0	50	117'000	160

3. Field testing

In most reported experiments dealing with occupational health effects of paving workers only pure bitumen is used as a binder. Limited data exist concerning the level of exposure and subsequent health effects when tar-containing binders are used. This is because tar binders are no longer used and are known to be dangerous. In Germany, even pure bitumen fumes are a matter of dispute and hence the bitumen producers are busy enough investigating the emissions of pure bitumen [16]. In Switzerland the low limits used in Germany and Austria regarding recycling asphalt are questioned, as they are not based on emission studies.

In the US, the National Institute for Occupational Safety and Health (NIOSH) and the Department of Transportation, Federal Highway Administration (FHWA) declared fumes a priority for non-regulatory action. Since 1997 a guideline produced by a cooperative effort between the asphalt industry, NIOSH, and FHWA advises the installation of exhaust ventilation systems at the auger area of the paver, where fumes are generated, for all new self-propelled HMA pavers weighing 16,000 pounds or more to reduce the impact of fumes for the roadmen [13].

3.1 Main emission sources and total emission

In order to determine the total emission in the paving process the main emission sources have to be considered. These were judged to be located as follows:

1. The material hopper, where the hot mix is filled in the paver
2. The auger and screed area of the paver, where the asphalt concrete is mixed, distributed and precompacted
3. The fresh pavement after the first compaction by the paver

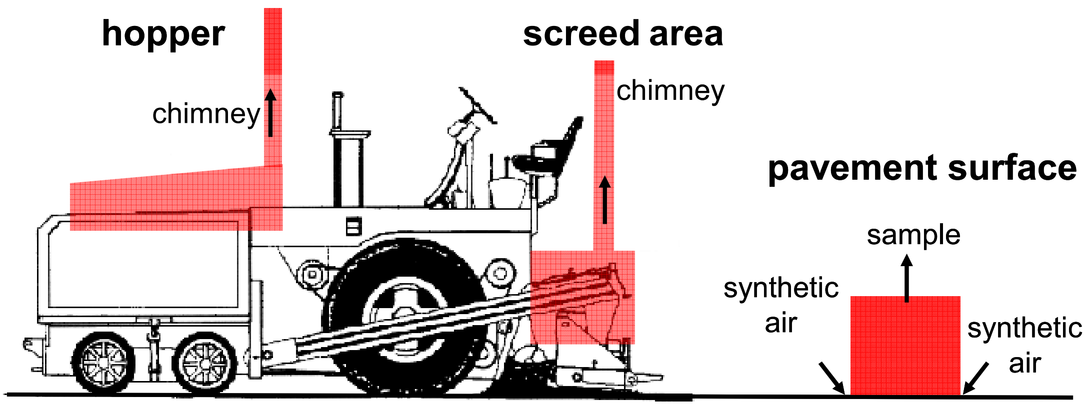

Figure 2: Enclosed paver and main emission sources

In our studies the three emission sources were measured separately. For this purpose the paver had to be adapted with removable enclosures over the material hopper and auger area, optimized by fume experiments. Two chimneys with a ventilator on the top were installed on the paving machine. Tubes connected the chimneys with the enclosed material hopper and the auger area. The flow through the chimneys was adjusted to 8 m^3/min each. Both chimneys were equipped with a set of instruments to measure temperature, air velocity and volume as well as sampling devices to collect PAH-emissions and particulate matter.

Emissions from the pavement surface were sampled with a modified Flex-cell of 150 mm diameter and a constant air flow of about 600 ml/min. The supplied air current was slightly higher than this to prevent the outside air from entering, because it could be contaminated by the exhaust fumes of the working machines.

Occupational health data were obtained from 7-8 paving workers equipped with personal samplers. Sampling was performed by deposition and adsorption on membrane filters in open-face filter holders, followed by PAH-adsorption tubes [15]. Four additional personal samplers were permanently fixed on the paver.

4. Results and discussion

4.1 Laboratory tests

PAH emission was expected to be a function of the PAH content in the binder. At first glance this is virtually true for the TPM as well as for Benzo(a)pyrene and the sum of 16 EPA-PAHs (Figure 3). A closer look reveals that this is not true anymore for TPM at PAH concentrations in the binder below 5000 ppm, where TPM emissions are nearly independent of the original PAH content.

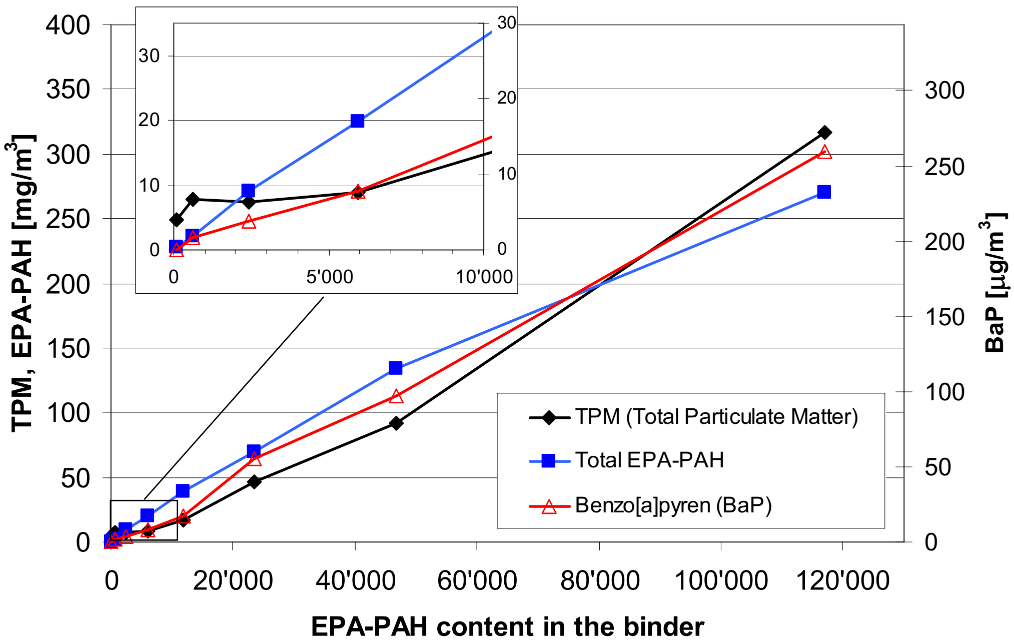

Figure 3: Emissions vs. PAH content in the binder

Temperature is the most important factor on the emissions as can be seen in Figure 4. TPM and Benzo(a)pyrene emissions increase almost exponentially with temperature. The temperature dependence can be approximated by the exponential function exp(1/T). This same function also describes the vapour pressure of individual chemical compounds. Emissions of TPM and low-volatile PAHs double with a temperature increase of approximately 12°C. This is consistent with the findings of Brandt who investigated the emission behaviour of different bitumen [8, 11]. However, the temperature dependence of the 16 EPA-PAHs is distinctly different. They show an almost linear and only modest increase with temperature. This is the effect of the rapid transfer into the gas phase of the low-volatile PAHs. At higher temperatures, and with increasing time, this leads to a significant depletion of the respective substances in the reaction vessel. Because low-volatile PAHs represent 90% of EPA-PAH in the emission, they dominate the behaviour of the cumulated PAHs.

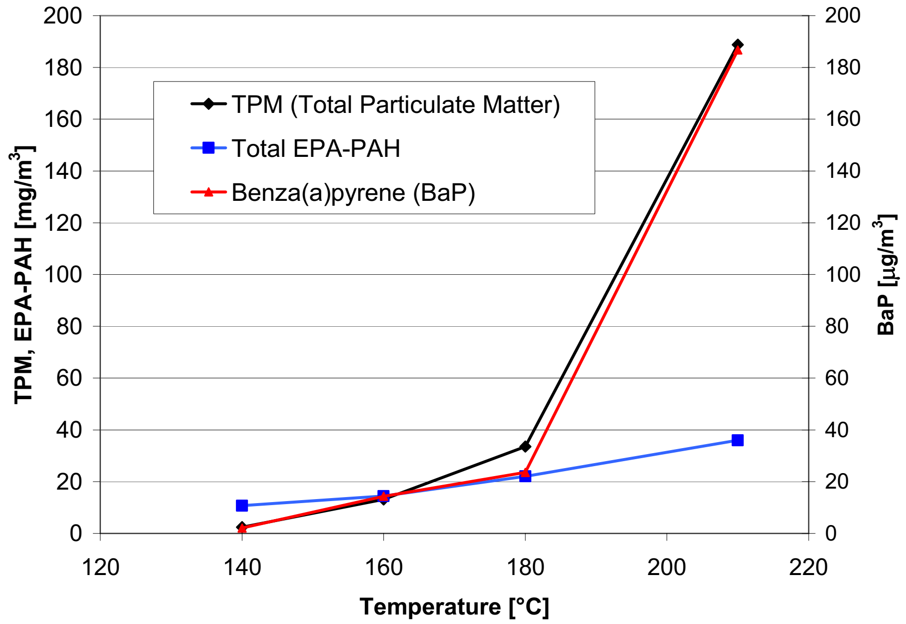

Figure 4: Temperature influence on the emission behaviour of a binder containing 5100 mg/kg EPA-PAH

4.2 Field tests

Three field tests were carried out in different parts of Switzerland. In the first field test the hot mix contained almost no tar, which can be seen at the low PAH-content of the extracted binder. The second field test took place in a tunnel, which is a special situation. As reported by the paving workers, ventilation was exceptionally good and a wind velocity of 0.7 m/s was measured. EPA-PAH-content of the paving material was slightly above the Swiss limit of 5000 mg/kg. In field test 3, where a bicycle lane was constructed, emissions were expected to be smaller as the lane is about half the width of a regular road lane.

Table 2: Field objects

	Field test 1	Field test 2	Field test 3
Location	national highway A2, breakdown lane	highway tunnel with 2-lanes	bicycle lane
Material	wearing course, asphalt concrete 32 mm with 30% RAP	wearing course with 65% RAP	asphalt concrete with about 50% RAP
Paving temperature	155±5°C	146±4°C	158±5°C
EPA-PAH-content in the binder	180±30 mg/kg	5600±300 mg/kg	2300 ±150 mg/kg

The scatter of the measured values from the personal samplers was larger than the difference between the results with and without coverage of the auger and the material hopper.

The capture efficiency corresponds to the fraction of the total emissions, coming from the material hopper and augers, which are evacuated through the ventilation. This capture efficiency was determined in the field using SF_6 as an inert tracer, which was dosed at a known rate, sampled in Tedlar bags and quantified using gas chromatography with an electron capture detector (GC-ECD). The so determined capture rate was 24 %, which is in contrast to the 80 % recovery rate demanded in the American guidelines. Our enclosure was a temporary and removable retrofit, which can not be expected to be as efficient as the specially designed pavers in the USA. Furthermore, the capture efficiency is determined indoors in the NIOSH Engineering guidelines, which will lead to more optimistic values. We, nevertheless, assume that even with excellent engineering it will be difficult to exceed 50 % capture efficiency in long term, rough field applications.

The distribution of the emissions to the presumed main sources can, nevertheless, be determined by taking into account the corresponding capture efficiency. In field test 3, a different paver was used which was, however, enclosed using the same technique. Some differences in the results might still originate from the different paver model.

Table 3: Main emission sources

Site	Parameter	emission [mass-%]		
		auger	material hopper	fresh pavement
Field test 2	TPM	84	10	6
	EPA-PAH	80	9	11
Field test 3	TPM	80	15	5
	EPA-PAH	55	25	20

Total mass flow was estimated from the measured concentrations at the three main sources and the corresponding air flow, taking into account the measured capture rate. The calculated mass flow amounted to 79 g/h and was clearly below the limit value of 500 g/h for particulates, defined in the Swiss regulation on air pollution [17]. In principal, this limit value is valid only for the emission of stationary sources, but it is used for diffuse or mobile emissions, such as the paving of road, as a guiding value.

The auger area is clearly identified as the main source with 55 to 85 percent of emissions (Table 1). The rest of the emissions come from the material hopper (9-25 %) and the pavement (5-20 %). In the case of the emitted fumes from the fresh pavement, there is a significant difference between the TPM and EPA-PAH-value. As the surface temperature of the compacted pavement decreases, only the volatile compounds are emitted. Because the low volatile compounds are passing through the filters and are collected only in the adsorption tubes, the TPM-value, which represents the lower and non-volatile compounds is lower at the colder sources than where the asphalt mix is hotter, which is at the auger and the material hopper.

4.3 Concentration of particulate matter

Particulate matter consists of the compounds which are retained by the filter. Mass determination is inexpensive and gives a rough estimate of the total amount of emissions and therefore, a limit value for TPM is fixed in many countries. Different countries, however, use slightly different measuring methods. The limit value for bitumen fumes is 10 mg/m^3 in Switzerland and Germany, and 5 mg/m^3 in the USA [18-20].

In the field tests the maximum TPM content was 3.8 mg/m^3 and hence below the limit value of 10 mg/m^3 (Table 3). TPM concentrations are strongly scattered, which is typical for outdoor emission measurements. In addition, paving workers work at different positions under changing fume concentrations. Concentrations in field test 3 are significantly lower than in field test 2. This may partly be due to the lower contamination of the RAP and the smaller lane width. However, the main factor is most likely the stronger crosswind, blowing the fumes away from the workers.

Table 4: Results of the field tests (nd: not determined)

	Field test 1	Field test 2	Field test 3
TPM concentrations of the personal samplers [mg/m^3]			
fixed on the paver	0.3 - 0.6	0.64 - 2.8	0.7 - 3.8
paving workers	nd	0.3 - 1.65	0.1 - 2.8
driver	nd	0.7 / 2.0	1.3 / 2.8
BaP concentrations of the personal samplers [µg/m^3]			
fixed on the paver	0.01 - 0.05	0.1 - 07	0.00 - 0.04
paving workers	nd	0.01 - 0.20	0.00 - 0.02
driver	nd	0.06 /0.11	0.03 / 0.07

5. Conclusions

Occupational health data obtained from the field tests in this research study show that emissions from RAP containing tar are all within the range of the Swiss limits of workplace concentrations [18]. However, the margin to maximum allowed values is not large, especially in the case of the measured TPM values. Furthermore, all three field tests took place under rather favourable conditions, because ventilation and wind velocities were always relatively high. Because of the large scatter of data, they should be verified with additional field tests.

It has been clearly demonstrated that the main emission source is the auger area, where the hot asphalt is mixed and distributed, but it has also become evident that the enclosure and capture of the emitted fumes from the auger area is difficult. In practice it would be extremely challenging to obtain a capture efficiency of 50 %, and even minimal ventilation is prone to somewhat hinder the paving process. Therefore, the enclosure of the paver is not recommended.

From the viewpoint of occupational health and based on the data of this study, the limit value of 5000 mg/kg EPA-PAH in the binder of the new hot mix, as fixed in the current guideline [2], is acceptable. This is true for paving temperatures below 160°C and sufficient ventilation or wind velocity. Still, the use of other techniques with lower paving temperature, especially cold recycling, should be encouraged for tar-containing RAP. In addition, the re-use of highly contaminated RAP with EPA-PAH binder contents above 20’000 mg/kg (approximately equivalent to a tar content of 10%) should be banned because the dilution of highly contaminated material is ecologically not advisable. RAP with an EPA-PAH content between 5000 and 20’000 mg/kg was added in concentrations up to 25% into the mixes resulting in EPA-PAH concentrations below 5000 mg/kg in the final mix. This dilution was explained by technical reasons; however, with the new generation of recycling plants, addition of 50% RAP and more is possible. Consequently, these moderately contaminated RAP cannot be used in these cases.

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