RESEARCH ARTICLE

Representativeness of the IAGOS airborne measurements in the lower troposphere

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In the framework of the In Service Aircraft for Global Observing System (IAGOS) program, airborne in-situ O_3 and CO measurements are performed routinely using in-service aircraft, providing vertical profiles from the surface to about 10–12 km. Due to the specificity of IAGOS measurements (measurements around busy international airports), uncertainties exist on their representativeness in the lower troposphere as they may be impacted by emissions related to airport activities and/or other aircraft. In this study, we thus investigate how the IAGOS measurements in the lower troposphere compare with nearby surface stations (from the local Air Quality monitoring network (AQN)) and more distant regional surface stations (from the Global Atmospheric Watch (GAW) network). The study focuses on Frankfurt but some results at other European airports (Vienna, Paris) are also discussed.

Results indicate that the IAGOS observations close to the surface do not appear to be strongly impacted by local emissions related to airport activities. In terms of mixing ratio distribution, seasonal variations and trends, the CO and O_3 mixing ratios measured by IAGOS in the first few hundred metres above the surface have similar characteristics to the mixing ratios measured at surrounding urban background stations. Higher in altitude, both the difference with data from the local AQN and the consistency with the GAW regional stations are higher, which indicates a larger representativeness of the IAGOS data. Despite few quantitative differences with Frankfurt, consistent results are obtained in the two other cities Vienna and Paris.

Based on 11 years of data (2002–2012), this study thus demonstrates that IAGOS observations in the lowest troposphere can be used as a complement to surface stations to study the air quality in/around the agglomeration, providing important information on the vertical distribution of pollution.

Keywords: airborne observations; representativeness; ozone; carbon monoxide; IAGOS; GAW

1 Introduction

In-situ observations are essential to improve our knowledge on the chemical composition of the atmosphere and its evolution with time, and to validate models and satellite observations. While the network of surface stations has considerably expanded over the last decades (although not in all regions of the world), in-situ observations still remain much sparser in altitude. Besides dedicated aircraft research campaigns that provide a detailed view of the chemical composition of the atmosphere but with a limited spatio-temporal coverage, routine in-situ measurements still essentially rely on radiosondes and commercial aircraft. Ozone (O_3) soundings began in the early 1960s and now provide a few thousand vertical profiles per year at several dozens stations throughout the world.

Routine in-situ airborne observations of chemical species not restricted to O_3 – e.g. carbon monoxide (CO), nitrogen oxides (NO_x), carbon dioxide (CO_2), methane (CH_4) – rely on only a few programs conducting airborne measurements using in-service aircraft since the 1990s. Among the programs still on-going, the oldest is the CONTRAIL (Comprehensive Observation Network for TRace gases by AIrLiner) program, within which greenhouse gases (CO_2, CH_4, N_2O, SF_6) measurements have been performed since 1993 using Boeing aircraft operated by Japan Airlines (Machida et al., 2008). Initiated in 1994, the MOZAIC (Measurement of OZone and water vapour by AIrbus in-service airCraft) program consisted in measuring O_3, CO and water vapour worldwide and routinely using as platform five in-service Airbus (A330/A340) long-haul aircraft

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operated by several international airlines (Marenco et al., 1998). In the framework of the CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) program, a much larger number of chemical species and physical parameters were measured from an instrumented cargo container installed on an Airbus aircraft from Lufthansa, for 4 flights per month (Brenninkmeijer et al., 2007). In 2005, MOZAIC and CARIBIC were joined in the preparatory phase of the European Research Infrastructure called IAGOS (In Service Aircraft for Global Observing System; Petzold et al., 2015) operational since 2011, which includes two branches: IAGOS-CORE (ex-MOZAIC) and IAGOS-CARIBIC. New instrumental developments were conducted in IAGOS-CORE and now allow the measurement of nitrogen oxides (NO_x) (on one aircraft from Lufthansa) (Berkes et al., in preparation) and in the near future (aeronautical certification of the instrument already approved) greenhouse gases (CO_2, CH_4) (Filges et al., 2015), while developments for aerosol measurements are still on-going (Bundke et al., 2015).

So far, many studies have used the IAGOS data to investigate air pollution throughout the troposphere, including the planetary boundary layer (PBL) (e.g. Ding et al., 2008; Elguindi et al., 2010; Kalabokas et al., 2007, 2013; Petetin et al., 2016a, 2016b; Sahu et al., 2014; Sauvage et al., 2005; Sheel et al., 2016; Tressol et al., 2008; Yamasoe et al., 2015). However, a part of the scientific community seems reluctant to use the IAGOS data for studying the lowermost PBL due to concerns about the representativeness of the IAGOS measurements close to the surface. While ozonesondes are usually launched in remote, rural or low-density urban areas, the specificity of the IAGOS measurements relies on the fact that the ascent/descent profiles are (i) slanted rather than strictly vertical, (ii) performed close to international airports (with potentially high emissions on the tarmac) (iii) located in the vicinity of large cities (among the 290 airports visited since 1994, the average population of the nearby city is 1.6 millions inhabitants), and (iv) along flight corridors frequented by other aircraft. For these reasons, questions arise on the representativeness of the IAGOS measurements in the lower troposphere (roughly, the 2 first kilometres). For instance, the Air Quality (AQ) community working on the validation of regional chemistry-transport models (thus focussing on the surface and the PBL) has sometimes argued that the IAGOS data are likely strongly influenced by very local pollution sources (e.g. airport, other aircraft) that cannot be correctly represented in the models with the current spatial resolution (a few kilometres). This would prevent relevant model-observation comparisons, in the same way that urban traffic surface stations are rejected for the validation of models at the surface (contrary to rural and urban background stations). However, the situation changed over the last years with model inter-comparison programs like AQMEII (Air Quality Model Evaluation International Initiative) or CAMS (Copernicus Atmospheric Monitoring Service) that benefited from the IAGOS dataset to evaluate models in the lower troposphere (e.g. Elguindi et al., 2010; Solazzo et al., 2013). Another example is given by the community working on the validation of CO tropospheric columns observed by satellite. Although several studies have used the IAGOS profiles to evaluate satellite products, mostly for CO (e.g. Clerbaux et al., 2008; Emmons et al., 2009; Worden et al., 2010), there are still concerns about the influence of these local pollution sources on the IAGOS observations close to the surface (where CO is the most concentrated) and thus on the pertinence of their use for the evaluation of satellite products (considering the fact that pixels have a typical size of a few tens of kilometres). Considering the scarcity of in-situ observations in altitude in the troposphere, there is therefore a strong need for a comprehensive study of the potential influence of the different local emissions (e.g. airport, other aircraft, nearby agglomeration) to assess the representativeness of IAGOS observations in the lower troposphere. This is required to better identify the type of applications allowed by the IAGOS dataset. Such analysis is of primary importance since, compared to the ~68,000 ozone soundings (since 1924) publicly available in the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) database, the IAGOS database now represents about 70,000 O_3 profiles since 1994 and 40,000 CO profiles since 2002 which, thus, contributes a large proportion of the whole dataset of in-situ profiles available in the troposphere.

Over the last two decades, several studies have investigated the consistency of the IAGOS O_3 dataset with radiosondes and surface stations (Thouret et al., 1998; Chevalier et al., 2007; Logan et al., 2012; Tilmes et al., 2012; Zbinden et al., 2013; Staufer et al., 2013, 2014; Tanimoto et al., 2015). The most recent and comprehensive study was made by Tanimoto et al. (2015), who compared collocated O_3 measurements (in space and time) obtained from IAGOS aircraft, ozonesondes and GAW (Global Atmospheric Watch) surface stations. Good correlations were obtained between surface stations and ozonesondes in the first 200 m (r = 0.8–1.0). Correlations were lower between surface stations and IAGOS (r ~ 0.6–0.8), likely due to the larger distance between sites compared with sondes (40–80 km), in agreement with Liu et al. (2009). These correlations were substantially improved after applying a simple wind selection (wind direction at $\pm 15°$ of the direction between the two locations, and wind speed above 2 m s^{-1}). In the first kilometre, a good consistency was found between the radiosondes and the IAGOS data profiles, with median relative differences around ±2%. Although this study greatly improved our confidence in the representativeness of the IAGOS O_3 measurements, a similar assessment of the representativeness of the IAGOS CO measurements is still missing. In addition, for the comparison between IAGOS and other datasets, some airports like Frankfurt (not taken into account in Tanimoto et al. (2015)) offer a much larger number of profiles, which can allow more in-depth analysis.

Although its secondary sources (oxidation of CH_4 and non-methanic volatile organic compounds (NMVOCs)) account for a large part of its global budget (around 57% of the total source (Stein et al., 2014)), CO is characterized by strong emissions associated with all types of incomplete combustion and a long lifetime (from a few weeks to a few months), which traditionally makes it a very useful tracer of pollution (Heald et al., 2003; Liu et al., 2003). Conversely, O_3 is a secondary pollutant, formed by a complex chemical mechanism involving notably NO_x, VOCs and CO, and likely to be quickly removed by surface dry deposition (Monks et al., 2015).

The overall objective of this study is to investigate more deeply the representativeness of the IAGOS O_3 and CO measurements in the lower troposphere. More specifically, this study aims at analysing how such airborne measurements are comparable to the surface observations measured by the nearby AQ monitoring network (AQN). Despite (usually) larger distances from the IAGOS airports, the comparisons will be extended to the GAW stations in order to assess how much the IAGOS observations throughout the PBL are influenced by the regional background. The subsequent objective of this study is to give recommendations on the possible use of the IAGOS dataset in the lower troposphere. The open research questions are: (i) are they relevant for the study of the AQ at the scale of the agglomeration? (ii) are they better suited for quantifying the impact of airborne activities on the local AQ? or (iii) are they able to provide information on the background pollution affecting the agglomeration?

The study will focus on Frankfurt, Germany, airport where IAGOS observations are the most abundant, but some insights from two other European airports at Vienna, Austria, and Paris, France, will be given. Both O_3 and CO are included. The analysis is done over 11 years between 2002 and 2012. The data and methods used in this study are described in Sect. 2. Results are analysed in Sect. 3, and summarized and discussed in Sect. 4.

2 Data and methods

2.1 IAGOS observations

This study mostly relies on the O_3 and CO observations available in the framework of the MOZAIC-IAGOS program (http://www.iagos.org) (Marenco et al., 1998; Petzold et al., 2015). Observations have been performed by commercial aircraft from several airlines since 1994 for O_3 and 2002 for CO. In both the MOZAIC and IAGOS programs, the same instruments are used in all aircraft. During the 2011–2014 overlapping years, inter-comparisons were systematically performed between the MOZAIC equipped aircraft and the IAGOS fleet, demonstrating a good consistency in the dataset (Nédélec et al., 2015). In MOZAIC, O_3 was measured using a dual-beam UV-absorption monitor (time resolution of 4 seconds) with an accuracy/precision estimated at about ±2 ppbv/±2% (Thouret et al., 1998), while CO was measured by an improved infrared filter correlation instrument (time resolution of 30 seconds) with an accuracy/precision estimated at ±5 ppbv/±5% (Nédélec et al., 2003). In IAGOS, both compounds are measured with instruments based on the same technology used for MOZAIC, with the same estimated accuracy and the same data quality control. A more detailed description of the IAGOS system and its validation can be found in Nédélec et al. (2015).

We use the barometric altitude available in the IAGOS database. It is deduced from the pressure measured by the aircraft, assuming standard conditions at the surface (temperature of 288.15 K, pressure of 1013.25 hPa). It is worth noting that this leads to an uncertainty on the actual altitude of the aircraft above the surface. In particular, under specific atmospheric conditions (cyclone), this barometric altitude can thus sometimes be located below the actual airport altitude. To tackle this uncertainty, it would be useful in the future to take into account the real conditions of temperature and pressure observed at the surface based on meteorological stations available in or around the airport. For now, such developments are yet not available and in this study, we will therefore consider all IAGOS measurements associated to their altitude above sea level (ASL), no matter if the altitude is below the actual elevation of the airport. Note also that the GPS altitude (measured by the aircraft system) is now available in the IAGOS dataset, but only since late 2014.

It is also worth noting that IAGOS profiles are not exactly vertical. The ascent/descent rates of IAGOS aircraft are 7.3 ± 2.0 m s^{-1} on average, i.e. 9 min to reach an altitude of 4 km. In parallel, the aircraft's horizontal speed (available in the IAGOS database) increases with altitude from about 85 m s^{-1} at 0–1 km to 166 m s^{-1} at 3–4 km on average. The horizontal distance between Frankfurt airport and IAGOS aircraft depending on the aircraft altitude is plotted in **Figure 1**. On average over the period 2002–2012, this leads to an horizontal displacement of IAGOS aircraft of about 9 ± 3, 19 ± 4, 36 ± 10, 41 ± 16, 71 ± 15 and 92 ± 17 km to reach an altitude of 0.5, 1, 2, 3, 4 and 5 km, respectively. Therefore, the IAGOS profiles have to be considered as semi-vertical profiles. However, it is worth noting that they can be considered as vertical profiles in model or satellite validation if the horizontal resolution (or the size of the pixel) is coarse enough (roughly, 1° for the 4 km above surface, 0.5° for the 2 km above surface, at mid-latitudes). For finer resolutions, the semi-verticality of IAGOS profiles has to be taken into account.

2.2 Surface observations

Surface O_3 and CO data from the local AQN are taken from the AIRBASE database (https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-7). AIRBASE data are available until January 2012. For comparison, some results in two other European cities, Vienna and Paris, will also be discussed. These two cities are chosen because they offer the largest number of IAGOS profiles (after Frankfurt) and a dense network of surface stations. For Paris, instead of AIRBASE, data are taken directly from AIRPARIF, the local agency in charge of the air quality monitoring network (https://www.airparif.asso.fr, data downloaded the 20th July 2017). In addition, we also use the CO and O_3 measurements available at GAW stations in West-Central Europe (see **Table 1**) in order to investigate the representativeness of IAGOS at a regional scale. GAW data are taken from the World Data Centre for Greenhouse Gases (WDCGG) (Schultz et al., 2015) (http://ds.data.jma.go.jp/gmd/wdcgg/; data downloaded the 1st of July 2016). Most of the GAW stations are located at less than 500 km from Frankfurt (the only exception being the Krvavec station 620 km away). At all these surface stations, O_3 is measured by UV absorption. The CO

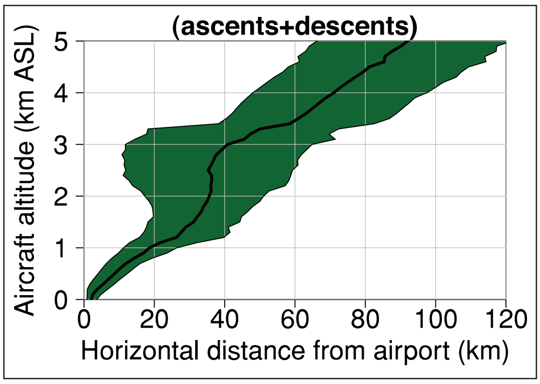

Figure 1: Horizontal distance between the Frankfurt airport and the aircraft depending on the aircraft altitude. The thick line indicates the mean distance, the green area delimits the range between 5th and 95th percentiles, over the period 2002–2012. Note that the offset distance (at the altitude of 0 m ASL) of a few kilometres roughly corresponds to the distance of the extremity of the airport runway. The Figure shows that the mean horizontal displacement of IAGOS aircraft in the lower troposphere ranges from 20 km at 1 km ASL to 90 km at 5 km ASL. DOI: https://doi.org/10.1525/elementa.280.f1

Table 1: Description of the surface stations from the GAW network. DOI: https://doi.org/10.1525/elementa.280.t1

Station name (contributor*)	Short name	Location	Elevation
Jungfraujoch (Empa)	JFJ	7.987°E, 46.548°N	3580 m
Sonnblick (EAA)	SNB	12.95°E, 47.05°N	3106 m
Krvavec (ARSO)	KVV	14.53°E, 46.30°N	1720 m
Schauinsland (UBA)	SSL	7.92°E, 47.92°N	1205 m
Rigi (Empa)	RIG	8.45°E, 47.06°N	1031 m
Hohenpeissenberg (DWD)	HPB	11.02°E, 47.8°N	985 m
Kosetice (CHMI)	KOS	15.08°E, 49.58°N	534 m
Payerne (Empa)	PAY	6.95°E, 46.82°N	490 m
Neuglobsow (UBA)	NGL	13.03°E, 53.17°N	65 m
Kollumerwaard (RIVM)	KMW	6.28°E, 53.33°N	0 m

* Contributors: DWD, Deutscher Wetterdienst; Empa, Swiss Federal Institute for Materials Science and Technology; RIVM, Dutch National Institute for Public Health and the Environment; UBA, Umwelt Bundesamt; EAA, Environment Agency Austria; CHMI, Czech HydroMeteorological Institute; ARSO, Environmental Agency of the Republic of Slovenia.

measurements usually rely on the techniques based on infrared absorption.

AQN include three main types of stations: (i) urban traffic stations located close to traffic emissions, usually at the kerbside, (ii) urban background stations located within the city or its suburbs (sometimes referred as suburban background stations) but further from the direct influence of traffic emissions (i.e. usually in a square or a small garden) with a typical representativeness ranging from a few km^2 to some tens km^2, and (iii) rural background stations located outside the city, and considered to have a representativeness of some hundreds of km^2. In this study, we will not distinguish urban background and suburban background stations, as differences among them are usually small compared with the two other types of stations. For clarity, a label is added to the name of each AQN station: "_R" for rural background, "_U" for urban background and "_T" for traffic.

Figure 2 shows the location of the GAW stations as well as the location of all surface stations around each airport. At Frankfurt (and nearby cities), the local AQN includes 16 surface stations: 1 rural background station, 9 urban background stations and 6 traffic stations. Note that due to several errors about the information relative to stations in AIRBASE, the type of station (e.g. traffic, urban/rural background) is identified directly from the German Federal Environmental Agency (Umwelt Bundesamt, http://www.env-it.de/stationen/public/stationList.do). Only 3 stations are located within the Frankfurt agglomeration, the other being located in nearby agglomerations: Mainz, Wiesbaden, Rüsselsheim and Darmstadt. At Vienna, 18 stations are considered, including 2 rural background stations, 12 urban background stations and 4 traffic stations. At Paris, 35 stations are taken into account, including 8 rural background stations, 21 urban background stations and 6 traffic stations. Information on the location and the altitude of all these surface stations are given in Table S-1 in the Supplement. Note that the same colours used in **Figure 2** to represent the different types of surface stations (green for GAW stations; purple,

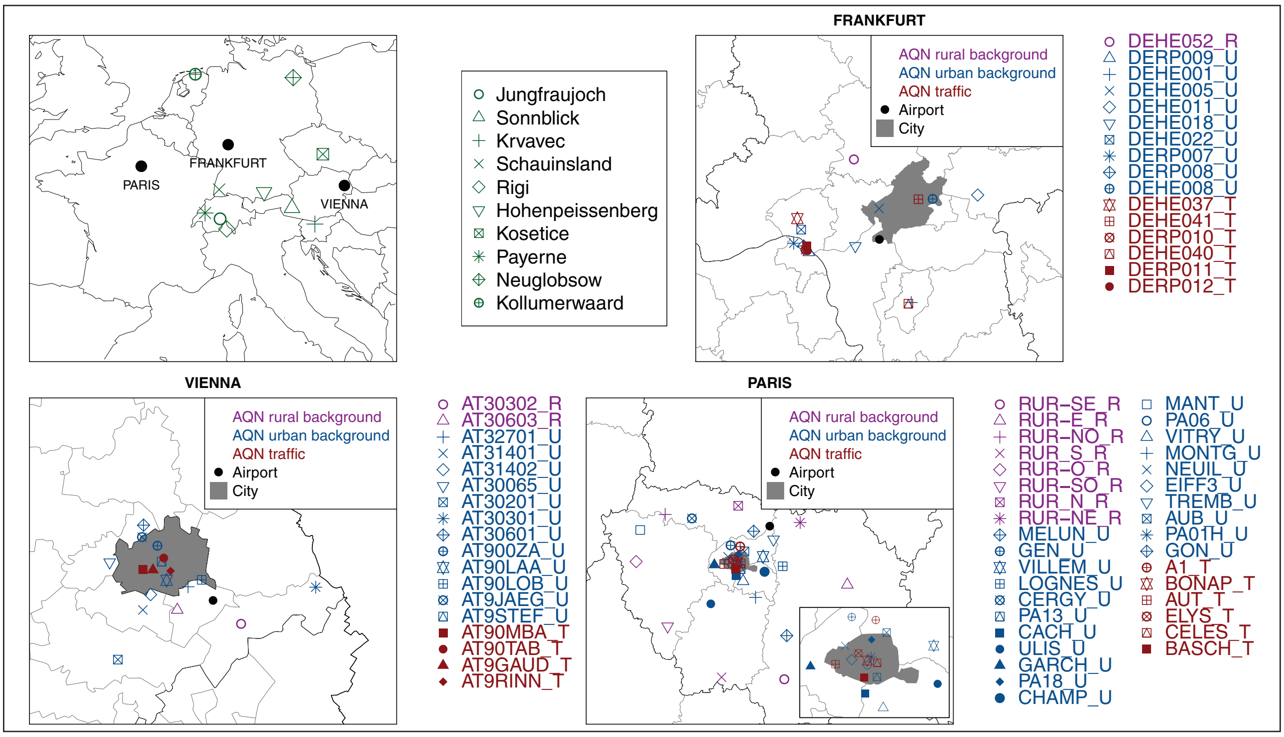

Figure 2: Location of airports and surface stations. The Figure displays a view of Europe with the 3 European airports and the GAW stations (top left panel) and a zoom on each city with the surface stations of the local air quality monitoring network (top right and bottom panels). Note that the grey area only delimits the borders of the city, not the borders of the agglomeration that extend further (in particular for Paris). DOI: https://doi.org/10.1525/elementa.280.f2

blue and red for rural background, urban background and traffic stations from the AQN, respectively) will be used in all the following figures.

2.3 Data treatment

Over the 2002–2012 period, a total of 12,569 ascent/descent profiles are available at Frankfurt airport (2,054 at Vienna and 1,610 at Paris). For information purposes, the location of IAGOS observations around airports is shown in the Supplement (Figures S-1 to S-3). In order to facilitate the comparisons with surface stations, IAGOS data are aggregated at the hourly scale over 50 m-deep layers from 0 to 4 km ASL from the raw IAGOS data (for instance, the "175 m" label will thus refer to the altitude layer ranging from 150 to 200 m ASL). At all surface stations, O_3 and CO mixing ratios are downloaded in the format of hourly averages. Surface stations give real hourly averages corresponding to the average of all quasi-instantaneous measurements of the instrument during a specific hour. However, it is worth noting that the hourly aggregates of IAGOS data do not correspond to hourly averages as one specific hourly value may be based on only one flight crossing the 50 m-deep layer in a few seconds, i.e. on only a few quasi-instantaneous observations. For convenience, for both airborne and surface data, we will refer indistinctly to hourly data in the paper but the reader should keep in mind that the IAGOS hourly averages are somewhat artificial.

The IAGOS measurements are sporadic in time, as they depend on the number of aircraft flying. Over the period 2002–2012, the mean number of ascent/descent profiles with available CO and/or O_3 data per day is 2 at Frankfurt airport (the maximum is 8 flights per day). Excluding the periods of technical failure in the system by considering only the days with at least one flight, this number increases to 3 flights per day on average. In terms of time of departure/arrival, the IAGOS flights at Frankfurt are mostly between 03:00 and 18:00 UTC, with almost no flight between 00:00 and 03:00 UTC. More details can be found in Petetin et al. (2016b). The comparison between IAGOS and surface data is made considering only the hours with simultaneous airborne and surface data (i.e. no time shift between them is allowed). Unless otherwise specified, all average CO and O_3 mixing ratios at surface stations discussed in this paper are calculated taking into account only the data when IAGOS flights took place. At a given hour, the availability of IAGOS data is not necessary the same at all levels (since for instance, an ascent profile may start at 14:59 at the surface and reach 4 km ASL at 15:08). Thus, surface data are considered when at least one IAGOS observation is available between 0 and 4 km ASL. Some comparisons between the two datasets are also made at the daily and monthly scale. For these analysis, hourly data are aggregated and averaged daily and monthly, without any minimum data coverage required. These daily and monthly data are used only for comparison, but do not correspond to real daily and monthly averages as they only include data sampled simultaneously by both aircraft and surface stations. Monthly time series of both CO and O_3 from each dataset are plotted in Figures S-4 to S-5 in the Supplement. Over the period 2002–2012 (4,018 days or 96,432 hours at total), the comparison is thus performed on 9% of the hourly dataset (8,348 hours). At daily and monthly scales, the availability of IAGOS data increases to 75 and 88%, respectively. A roughly similar amount of IAGOS data is available during all four seasons (for O_3: 24% in winter, 20% in spring and summer, 23% in autumn).

3 Results

3.1 Overall distribution of mixing ratios

The distribution of CO and O_3 hourly mixing ratios over the period 2002–2012 is shown in **Figure 3** for all surface stations and a subset of IAGOS altitude levels below 4 km ASL. In terms of CO pollution, a clear distinction among

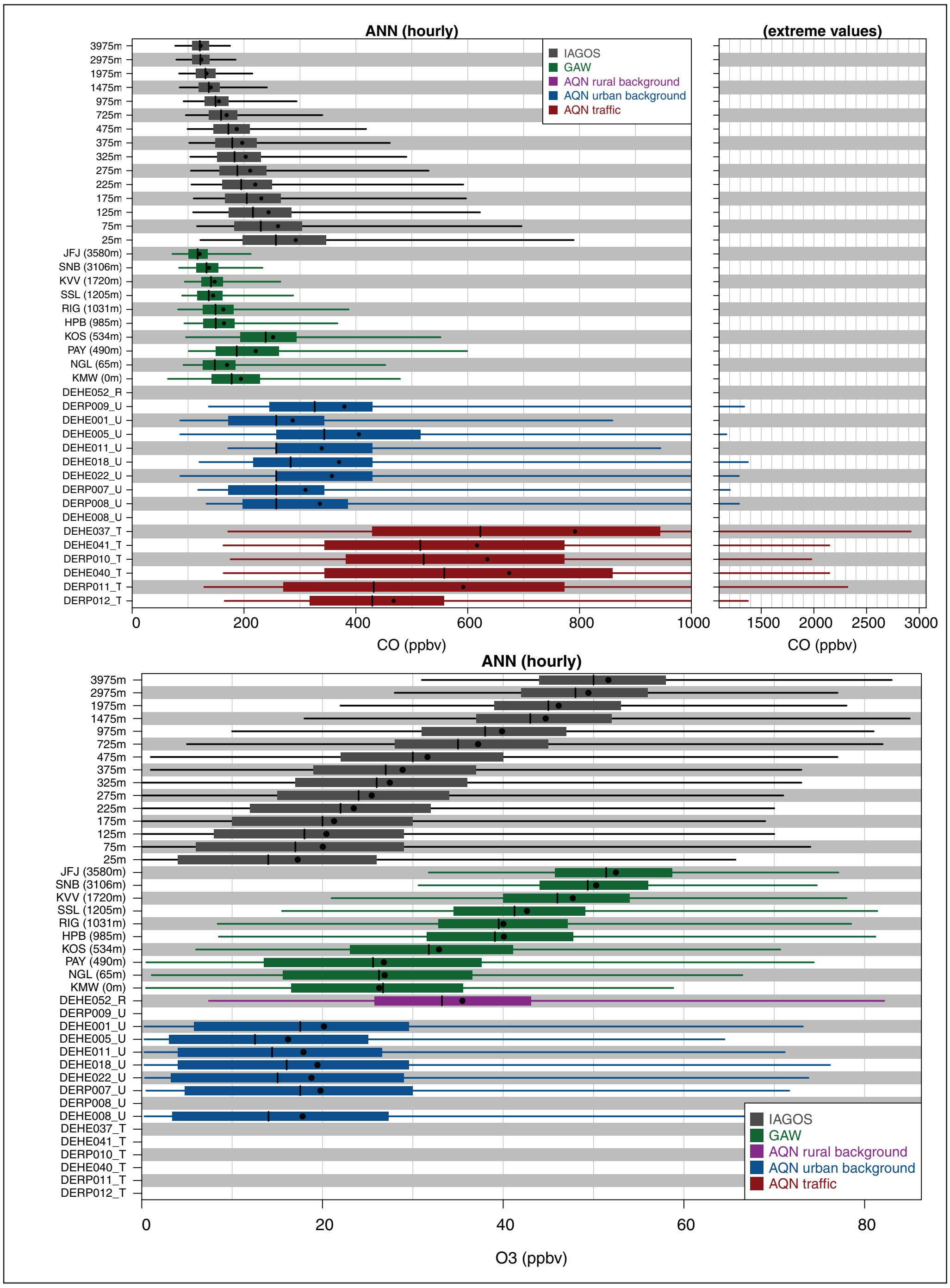

Figure 3: Distribution of the CO and O_3 hourly mixing ratios at Frankfurt, over the period 2002–2012. The Figure displays the 1st, 25th, 50th, 75th and 99th percentiles (box-and-whisker) and the mean (black point). For CO, a secondary plot (on the right) shows the highest 99th percentiles with a different scale on the abscissa. The "125 m" label in IAGOS data refers to the 50 m-deep layer ranging from 100 to 150 m ASL. A specific colour is used for each type of stations and for IAGOS. Close to the surface, IAGOS data show CO and O_3 mixing ratios in reasonable agreement with urban background stations. The CO mixing ratios measured by IAGOS are much lower than the mixing ratios measured at traffic stations. Higher in altitude, a growing consistency with GAW regional stations is highlighted. DOI: https://doi.org/10.1525/elementa.280.f3

the 3 different types of surface stations is highlighted. On average, the CO mixing ratio is 630 ± 414 ppbv (mean standard deviation of the different stations) at traffic stations, 348 ± 212 ppbv at urban background stations, and 171 ± 61 ppbv at GAW stations. As previously mentioned (see Sect. 2.3), these averages are calculated based on the hourly surface data with concomitant IAGOS flights. Such differences between the different types of stations are expected considering the increasing distance to strong emission sources like traffic. At the 3 first altitude levels (0–50, 50–100, 100–150 m ASL), the IAGOS measurements give a CO mixing ratio of 292 ± 142, 261 ± 124 and 244 ± 109 ppbv, respectively. This is the first important result: at Frankfurt, even at the lowest altitude levels, the CO mixing ratios measured by IAGOS aircraft never reach mixing ratios as high as measured at traffic stations, but roughly correspond to the urban background pollution measured in the nearby cities and are even slightly lower by 16, 25 and 30% in these 3 first levels, respectively. We calculated the relative bias of IAGOS versus surface stations from the local AQN considering simultaneously all individual hourly data from surface stations with concomitant IAGOS data at the corresponding altitudes (i.e. the station elevation). The overall bias on CO reaches –26% for urban background stations, and –61% for traffic stations (more details on this bias are given below in Sect. 3.2).

As mentioned in Sect. 2.3, contrary to surface stations, the IAGOS data used here are based on hourly aggregates of sporadic (quasi-)instantaneous observations. Compared with surface data, its temporal variability (represented by the standard deviation) may thus be biased high since it includes not only the sub-hourly variability but also a part of the instrument noise (smoothed in the surface hourly averages). Despite that, IAGOS shows close to the surface a temporal variability much lower than the variability observed at the surface stations (by a factor of 1.5–2 in the 3 first altitude levels). The second important result is that the CO mixing ratios quickly decrease with altitude, and reach the average mixing ratio of GAW stations (171 ± 61 ppbv) at 650–700 m ASL. A similar picture is obtained at the seasonal scale (not shown). Interestingly, the altitude levels at which IAGOS reaches the average CO given by the GAW stations slightly varies with the season, from about 650 m in winter/autumn to 750 m in spring/summer. This is consistent with the seasonal variations of the PBL height. More details on the comparison between IAGOS and GAW are given below in Sect. 3.3.

The distribution of hourly O_3 mixing ratios is shown in **Figure 3** (bottom panel). Note that due to the fast titration of O_3 by the NO emitted by car engines, traffic stations usually do not measure O_3. The mean O_3 mixing ratio is 19 ± 17 ppbv at the urban background stations and 35 ± 15 ppbv at the (unique) rural background station of the AQN. For the GAW stations, mixing ratios range between 26 ± 13 ppbv at lowest elevations (KMW, 0 m) and 52 ± 10 ppbv at the highest (JFJ, 3580 m). The mean O_3 mixing ratios observed by IAGOS in the first 3 altitude levels are 17 ± 16, 20 ± 17 and 20 ± 16 ppbv, respectively. The corresponding relative differences with the mean urban background O_3 mixing ratios are thus –7, +8 and +10%. Comparing all individual hourly data with concomitant IAGOS data at the corresponding altitudes, the mean difference between IAGOS and surface O_3 is +3% for urban background stations. Thus, as for CO, the O_3 measured by IAGOS aircraft close to the surface appears close to the urban background measured by surface stations. The mean O_3 measured at the rural background station DEHE052_R is much higher (35 ± 15 ppbv), but this is due to its location in the Taunus observatory at the summit of Kleiner Feldberg (elevation of 811 m), as discussed in Sect. 3.3. At this altitude, IAGOS shows O_3 mixing ratios only 12% higher than DEHE052_R, thus in reasonable agreement.

3.2 Diurnal variability

As mentioned in Sect. 3.1, the exact comparison between IAGOS and urban background stations gives small differences. Reasonable correlations are also found between the two datasets (r = 0.58 and 0.81 for CO and O_3, respectively). In this section, we investigate the diurnal variability of these differences. The biases between the two datasets are calculated for each hour of the day, and are plotted in **Figure 4**. Here, the bias designates IAGOS minus the urban background stations. These biases show a substantial diurnal variability. Both the negative bias of CO and the positive bias of O_3 at urban background stations are greater (in absolute value) during the morning (06:00–09:00 local time (LT = UTC+1)) when they reach –40% for CO and +70% for O_3. After 10:00 LT, these biases get smaller until a minimum is reached during afternoon, when they range between ±20% for both CO and O_3. They increase again in late afternoon (up to ±40%). For O_3, the bias is not significant during the afternoon. The correlation follows similar diurnal variations with a minimum during the early morning (0.4–0.6) and a maximum during the afternoon/evening (0.7–0.9).

In these comparisons, we considered the IAGOS data at the altitudes corresponding to the elevations of the urban background stations, which range between ±50 m around the elevation of the Frankfurt airport (see the zoom panel in **Figure 5**). Thus, even during nighttime, early morning or late afternoon, most of these IAGOS observations likely still belong to the boundary layer (i.e. we are not comparing observations at the surface with observations in the nocturnal residual layer). Therefore, the diurnal variations of the differences between IAGOS and urban background stations, and more specifically the stronger discrepancies during early morning and late afternoon, are likely more explained by the combined effect of higher local emissions and less dispersive conditions. Indeed, these times of the day correspond to the well-known morning and evening peaks of traffic emissions (rush hours). Although these urban background stations are not located at the kerbside like traffic stations, they are still greatly influenced by traffic emissions that represent one of the major sources of pollution, as illustrated by the diurnal variations of CO or NO_x (not shown). These traffic emissions increase the CO mixing ratios (more emissions)

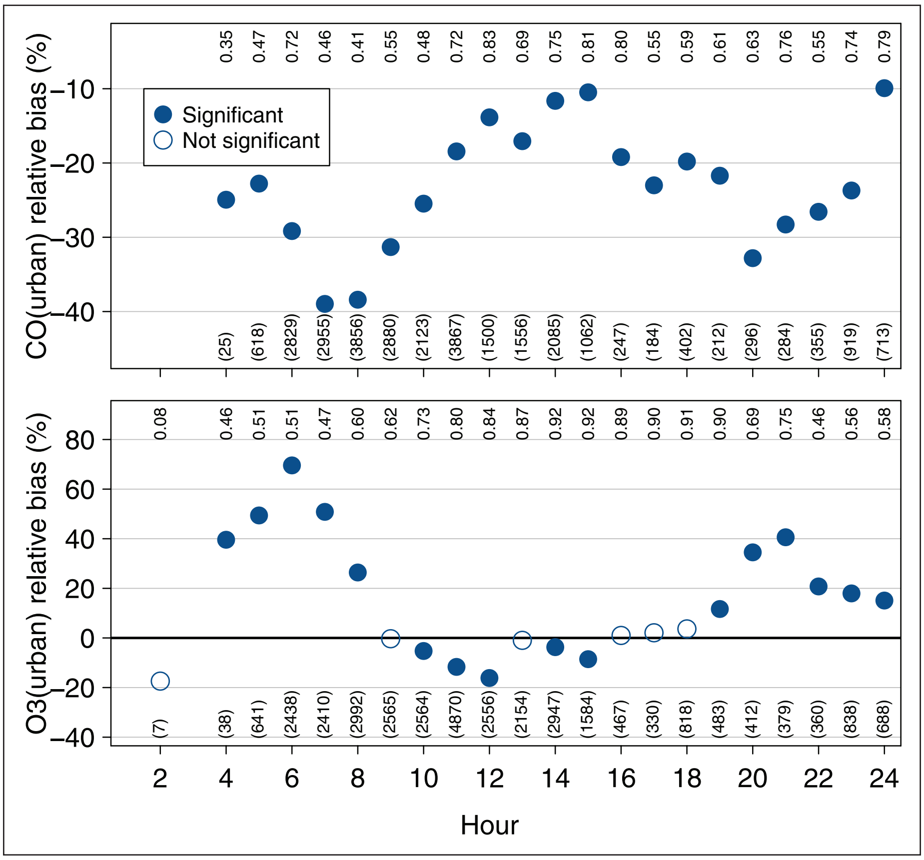

Figure 4: Diurnal variations of the relative bias between IAGOS and urban background stations. Results are shown for CO (top panel) and O_3 (bottom panel) at Frankfurt. Hours are in LT. The filled circles indicate a statistically significant difference (at a 95% confidence level) while the empty circles indicate that the difference is not significant (too low number of points and/or too small difference). Both the correlation (r, at the top of each panel) and the number of points included in the calculation (at the bottom of the panel, in brackets) are reported on the Figure. For both CO and O_3, the bias between IAGOS and urban background stations is found to be higher during the morning and early evening, and lower during the afternoon. DOI: https://doi.org/10.1525/elementa.280.f4

and decrease the O_3 (through a stronger titration by the NO emitted by the vehicles). In parallel, the PBL is not fully developed and the (horizontal and vertical) mixing within the PBL still moderate, which may hinder the dispersion of the local pollution.

Note that focusing only on the 12:00–17:00 LT time window, the mean difference between IAGOS and urban background stations reaches −14 and –4% for CO and O_3, respectively (compared to –26 and +3% when all data are taken into account). Since we are interested in investigating the representativeness of the whole IAGOS dataset, we will keep considering all data whatever the time of the day in the rest of the paper, although some insights during the 12:00–17:00 LT time window will be given for information purpose.

3.3 Vertical distribution

The CO mixing ratio is plotted against altitude for IAGOS measurements and elevation of the surface stations in the lowest troposphere in **Figure 5** (top panel). Due to differences of elevation (from 0 m at KMW to 3,580 m at JFJ) and nearby environment (e.g. site in low-density urban area, coastal site, mountain site), the CO pollution varies strongly from one GAW station to the other. Substantial differences between IAGOS and GAW data are observed below 1 km. Compared to IAGOS, the two GAW stations at 500 m (PAY and KOS) show higher CO while the two stations close to 0 m (KMW and NGL) show lower CO mixing ratios. The mixing ratios at all other GAW stations closely follow the IAGOS vertical profile at Frankfurt. A good consistency between IAGOS and GAW is also found during all 4 seasons (not shown). Both IAGOS and GAW data tend to show a decrease of CO with altitude close to the ground. A decrease of CO with altitude is also observed among urban background stations. This feature is not observed at the traffic stations, due to the predominant contribution of the traffic emissions. As discussed in Sect. 3.1, the CO mixing ratios measured by IAGOS are slightly lower than the urban background, but much lower than the CO mixing ratios at traffic stations. Concerning the variations of O_3 with altitude (**Figure 5**, bottom panel), a very good agreement is found between IAGOS and the different surface stations, whatever their elevation, although some discrepancies persist for the lowest GAW stations. These results are consistent with the study of Chevalier et al. (2007) that highlighted over the period 2001–2004 a good coherence between IAGOS O_3 vertical profiles and

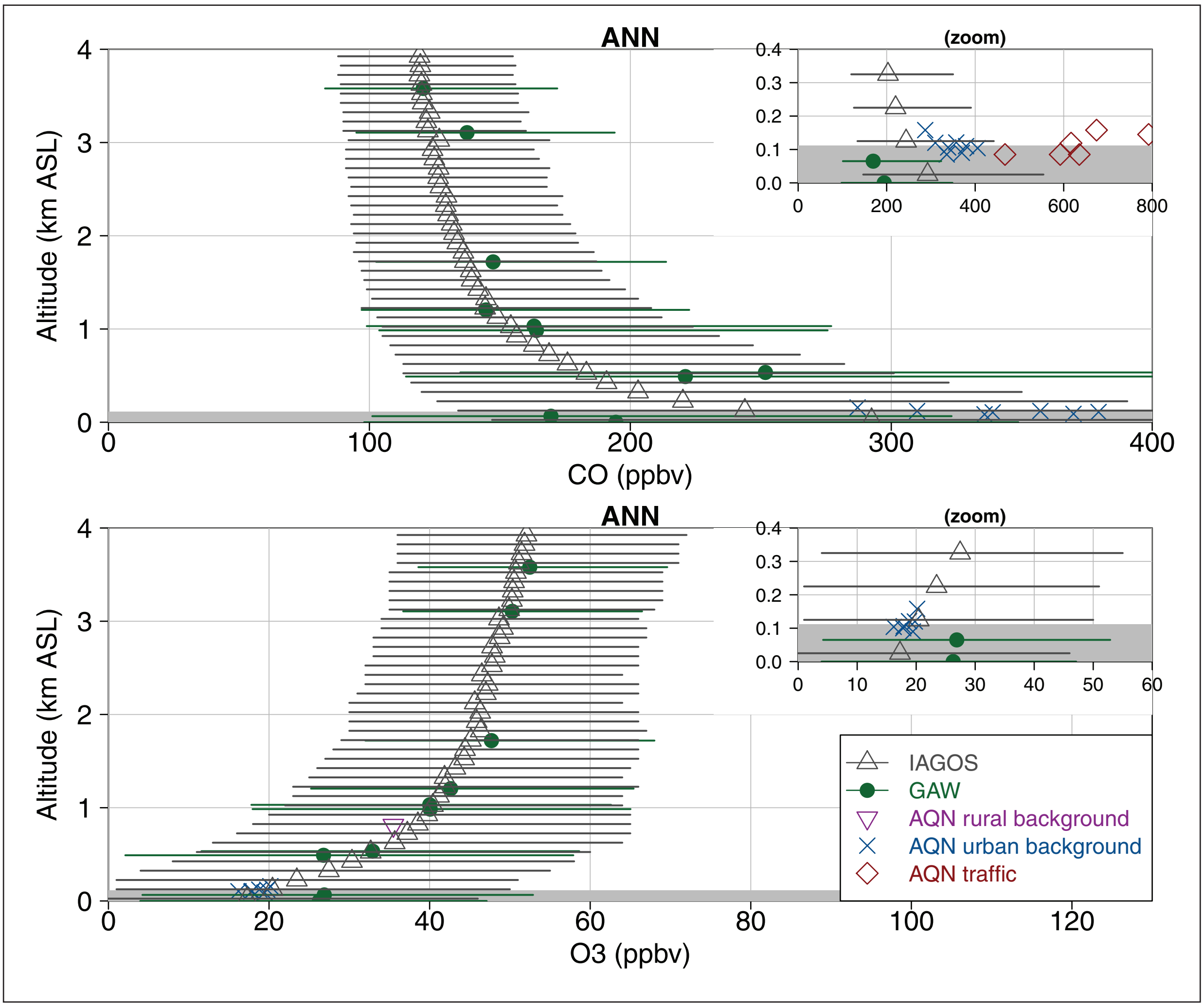

Figure 5: CO (top panel) and O_3 (bottom panel) mixing ratios versus altitude. This Figure includes observations from IAGOS aircraft, surface stations at Frankfurt and GAW stations. The grey area indicates the elevation of the Frankfurt airport (110 m). As previously mentioned (Sect. 2.1), the altitude of IAGOS measurements is deduced from the pressure assuming standard conditions at the surface, which explains that some points may have a barometric altitude below the actual airport elevation (e.g. presence of strong low-pressure systems). Both the mean (points) and the 5th and 95th percentiles (horizontal lines; only for IAGOS and GAW) are shown. A good agreement between IAGOS and all surface stations is found, except for traffic and some GAW stations (below 500 m ASL) for CO. DOI: https://doi.org/10.1525/elementa.280.f5

a larger set of surface stations in Europe, but with larger differences at lower altitudes.

3.4 Seasonal variations

The mean seasonal variations of CO and O_3 at the different surface stations and IAGOS altitude levels are shown in **Figure 6**. In terms of mixing ratios, the distinction between the different types of surface stations is again obvious and persists all year round. All surface stations and IAGOS altitude levels show similar seasonal variations with maximum CO in winter, and minimum CO in summer. However, results highlight differences of amplitude in the seasonal cycle. Traffic and urban background stations show a mean relative amplitude (here defined as the maximum minus minimum of the seasonal cycle normalized by the annual average) of 52 ± 15 and 68 ± 4%, respectively. At these stations, the amplitude tends to decrease as the mean CO mixing ratio is increasing. At GAW stations, the relative amplitudes range between 36 to 79%, the lower amplitudes being usually found at higher elevations. A notable exception is the KMW station (0 m ASL) that shows a relative amplitude of only 42%, i.e. comparable to the relative amplitude observed at the highest mountain stations. Concerning IAGOS, the amplitude is the highest (50–55%) in the first 500 m, and quickly decreases with altitude down to 30–35% above 1 km ASL. The bias between IAGOS and urban background stations is found to vary with the season, from about –30% in winter/autumn to –14% in summer (against –26% at the annual scale, see Sect. 3.1). Compared with GAW stations, IAGOS (at the corresponding altitude) usually shows a slightly lower CO relative amplitude (the relative bias of amplitude is –20% on average over all GAW stations), mainly due to slightly lower CO during wintertime (–8%).

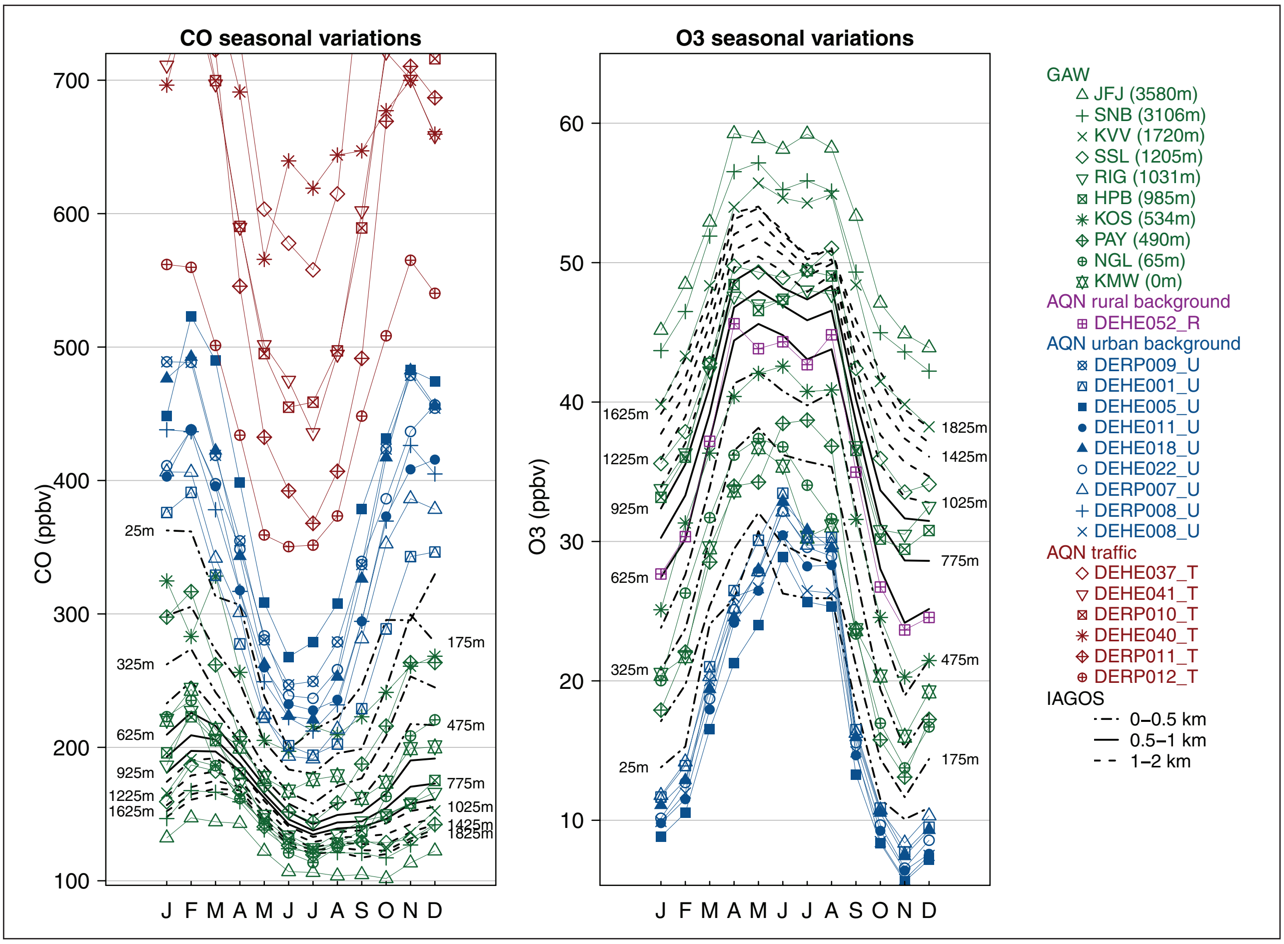

Figure 6: Average monthly variations of CO and O_3 mixing ratios. This Figure includes observations from IAGOS aircraft, surface stations at Frankfurt and GAW stations. For clarity, the IAGOS monthly variations are represented at several (50 m-deep) levels (one black line per 50 m-deep layer, every 3 layers, i.e. 25 m, 175 m, 325 m, etc.) gathered in three groups: dot-dashed, solid and dashed lines for altitude levels ranging between 0–0.5, 0.5–1 and 1–2 km ASL, respectively. The maximum CO mixing ratios at traffic stations in winter are below 850 ppbv. The Figure shows that the differences among the different types of stations persist all year round. IAGOS data show lower CO in winter, leading to a lower amplitude than surface stations. DOI: https://doi.org/10.1525/elementa.280.f6

For O_3, a common seasonal pattern is observed at the surface stations and at altitude, with a broad spring-summer maximum and a minimum in late autumn-early winter. This is in good agreement with other observations in the lower troposphere (Parrish et al., 2013). As for CO, the relative amplitude of the IAGOS dataset quickly decreases with altitude from about 100% close to the surface to 30–40% above 1 km ASL. The relative amplitudes observed at the GAW stations are in close agreement with IAGOS at the corresponding altitude levels (relative mean bias of amplitude of –1%). However, the O_3 relative amplitude observed at the urban background stations range between 121–144% (134% on average) and is thus slightly stronger compared to what is observed both by aircraft and GAW stations at these altitudes. This is mainly due to a lower minimum of O_3 in winter at the urban background stations. The absolute amplitude (maximum minus minimum) is quite similar to the other datasets. As for CO, the bias on O_3 between IAGOS and urban background stations thus depicts some seasonal variations, from +28% in winter to –12% in summer (against +3% at the annual scale, see Sect. 3.1). These higher differences for both CO and O_3 in winter could be due to less dispersive conditions during the cold season.

3.5 Temporal variability

We now investigate how the CO and O_3 temporal variability observed in IAGOS airborne measurements is comparable to the variability at the surface. The Pearson correlation coefficients (r) between the surface stations and the different IAGOS altitude levels are shown in **Figure 7**. These correlations are calculated based on hourly, daily and monthly aggregates.

For CO, the correlations between hourly time series of IAGOS and AQN stations are highest close to the surface (r ~ 0.5–0.7 for urban background stations, r ~ 0.4–0.6 for traffic stations) and decrease with altitude (almost no correlation above 2 km ASL). A different picture is found with the GAW stations. For most GAW stations, the correlation with IAGOS hourly time series is low close to the surface and increases with altitude up to a maximum. Interestingly, the altitude at which this maximum

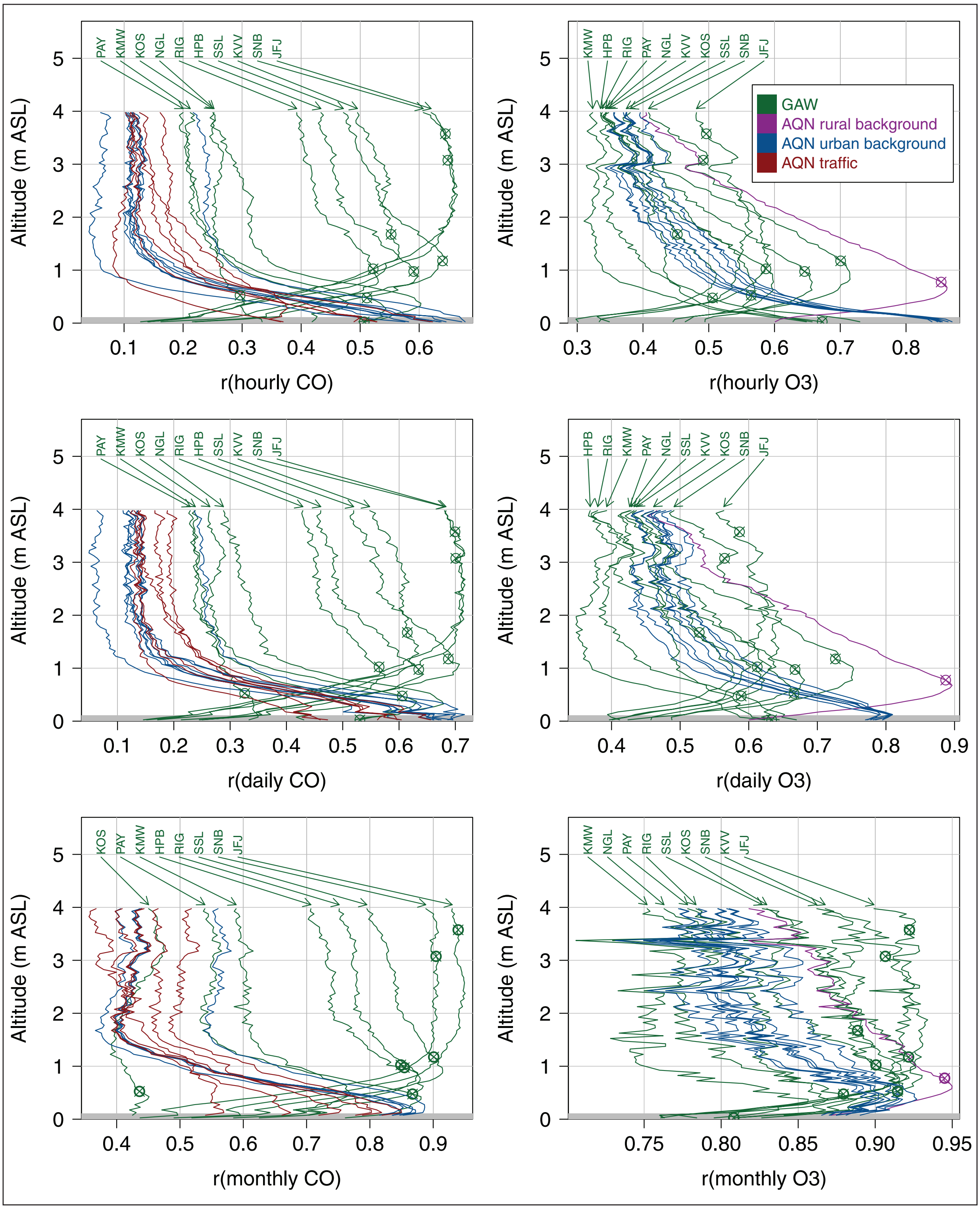

Figure 7: Profiles of correlation between the IAGOS and surface stations. Profiles are shown for the hourly (top panels), daily (middle panels) and monthly (bottom panels) data at Frankfurt, for CO (left panels) and O_3 (right panels). For clarity, only the name of GAW stations is written. For each GAW station and for the rural background station, the elevation is indicated on the corresponding curves by open circles with a cross. The altitude of the Frankfurt airport is shown in grey. The correlation between IAGOS and AQN traffic and urban background stations is the highest close to the surface and decreases with altitude. Conversely, the correlations between IAGOS and GAW is low to moderate close to the surface, increases with altitude up to a maximum and then decreases. The altitude of maximum correlation roughly corresponds to the elevation of the GAW stations. DOI: https://doi.org/10.1525/elementa.280.f7

correlation is reached is relatively close to the elevation of the corresponding surface station. To highlight this feature, open circles with crosses are added in **Figure 7** at the elevation of the corresponding stations (only for both GAW stations and the rural background station). For instance, the correlation between IAGOS and SSL

(HPB) – elevation of 1,205 m (985 m) – is 0.16 (0.22) at the lowest altitude level and increases up to 0.64 (0.60) at 1,325 m (875 m) ASL. The main exception is the KOS station that is poorly correlated with IAGOS data whatever the altitude. This result indicates that at a given altitude, the CO variability is reasonably homogeneous at the regional scale in this part of Europe. It is worth noting that these low to moderate correlations are expected at the hourly scale, as we are comparing data sampled at quite distant locations. When considering daily and monthly time series, the correlations are all strongly enhanced but the shape of the profiles remains the same. Note that the high correlations observed at the monthly scale are driven by the seasonal variations. Considering deseasonalized monthly time series, we obtained correlations roughly comprised in the range between the previous daily and monthly correlations (not shown). Thus, this is another important result: the IAGOS observations in the lowest troposphere around Frankfurt share a large part of their variability with the nearby surface stations, likely due to common processes driving CO mixing ratios (e.g. local emissions and meteorology including PBL dynamics). However, as one moves higher in altitude, the variability of IAGOS quickly deviates from the local variability to move closer to the regional-scale variability partly driven by mesoscale meteorology such as frontal passages.

The correlations between O_3 hourly time series from IAGOS and AQN urban background stations are strong close to the surface (around 0.85) and decrease with altitude (down to 0.3–0.4 at 4 km ASL). Between IAGOS and GAW stations, the correlation profiles highlight some common features with CO, namely an increase of the correlation with altitude up to a maximum. The altitude of maximum correlation also appears to be related to the station elevation, but the discrepancies between both can be stronger than for CO. For instance, the maximum correlation between IAGOS and SNB (elevation of 3,106 m) is reached at about 1,500 m ASL. The agreement for the AQN rural background station is relatively good (maximum correlation at about 600–700 m ASL for an elevation of 800 m). As for CO, the correlation is higher when considering daily or monthly time series and the results remain qualitatively the same.

Note that when considering only the 12:00–17:00 LT time period, the picture remains the same except that correlations between IAGOS and AQN stations increase to 0.6–0.8 for CO and 0.9 for O_3 at the hourly scale (as expected, see **Figure 4** and Sect. 3.2). Note also that although some correlations between GAW and IAGOS data are relatively low to moderate for both CO and O_3, they remain in the range of the correlations obtained between GAW stations themselves. To illustrate it, for both CO and O_3, the correlations between all pairs of GAW stations have been calculated based on hourly time series. They are plotted on **Figure 8** (green triangles) as a function of the difference of elevation (in absolute value) between two given GAW stations. Similarly, we also plotted on **Figure 8** the correlation between GAW stations and IAGOS observations at each altitude level (i.e. one grey point in **Figure 8** represents the correlation between the hourly time series at one given GAW station and the hourly time series of IAGOS data at one given altitude level) against the difference of altitude (i.e. the difference between the elevation of the GAW station and the IAGOS altitude considered). Correlations are usually the lowest for biggest altitude differences, but increase as the difference of altitude gets smaller. At similar altitudes (differences below a few hundreds of metres), the correlations obtained are quite variable but tend to be higher.

3.6 Trends

We now investigate how trends deduced from IAGOS measurements are comparable with the trends observed at the surface stations. Annual trends calculated by linear regression based on the time series of CO annual averages are shown in **Figure 9**. Most trends show a statistically significant decrease (at a 95% confidence level) of CO over the period 2002–2012. In agreement with numerous other studies (e.g. Dils et al., 2011; Worden et al., 2013; Petetin et al., 2016a), this decrease over Europe is mainly due to the reduction of CO anthropogenic emissions over the last decades (Yoon and Pozzer, 2014; Strode et al., 2016). The strongest decrease is observed at the traffic stations with best estimates of the trend ranging from –5.5 to –10% yr^{-1}. The urban background stations show lower negative trends, ranging from –2.5 to –6% yr^{-1}. Such differences of CO trends between traffic and urban background stations have already been highlighted in Europe by Guerreiro et al. (2014). Among the GAW stations, 7 stations have a negative trend ranging between –4 and –1% yr^{-1} (JFJ, SNB, SSL, RIG, PAY, NGL, KMW), while the trends at the 3 other stations are not significant (KVV, HPB, KOS). In the lowest altitude levels, IAGOS data show a significant decrease of CO mixing ratios at the same rate as urban background stations (around –4% yr^{-1}). Contrary to most traffic stations, the difference of trends between IAGOS and urban background stations are usually statistically insignificant. The IAGOS negative trends decrease with altitude, down to –2% yr^{-1} at 2 km. Such a decrease of CO in the IAGOS dataset of Frankfurt has already been highlighted by Petetin et al. (2016a). Thus, as for the distribution of CO mixing ratios, IAGOS observations progressively shift from a behaviour similar to urban background stations to a behaviour in reasonable agreement with the highest GAW stations, as one moves toward higher altitudes. Quite similar features are observed with seasonal trends (not shown).

All O_3 annual trends from IAGOS and the AQN appear statistically not significant (see Figure S-6 in the Supplement). No significant differences between these two datasets are highlighted. Among the GAW stations, 2 stations (KOS and NGL) out of 10 show a significant decrease of O_3 over the period 2002–2012, although these trends are not strongly significant (about -1.5 ± 1.2% yr^{-1}). They are mostly driven by a significant decrease in summer and/or autumn. Slightly negative or insignificant trends of O_3 in Europe over the 2000s have already been highlighted in other studies (e.g. Cui et al., 2011; Logan et al., 2012; Parrish et al., 2014).

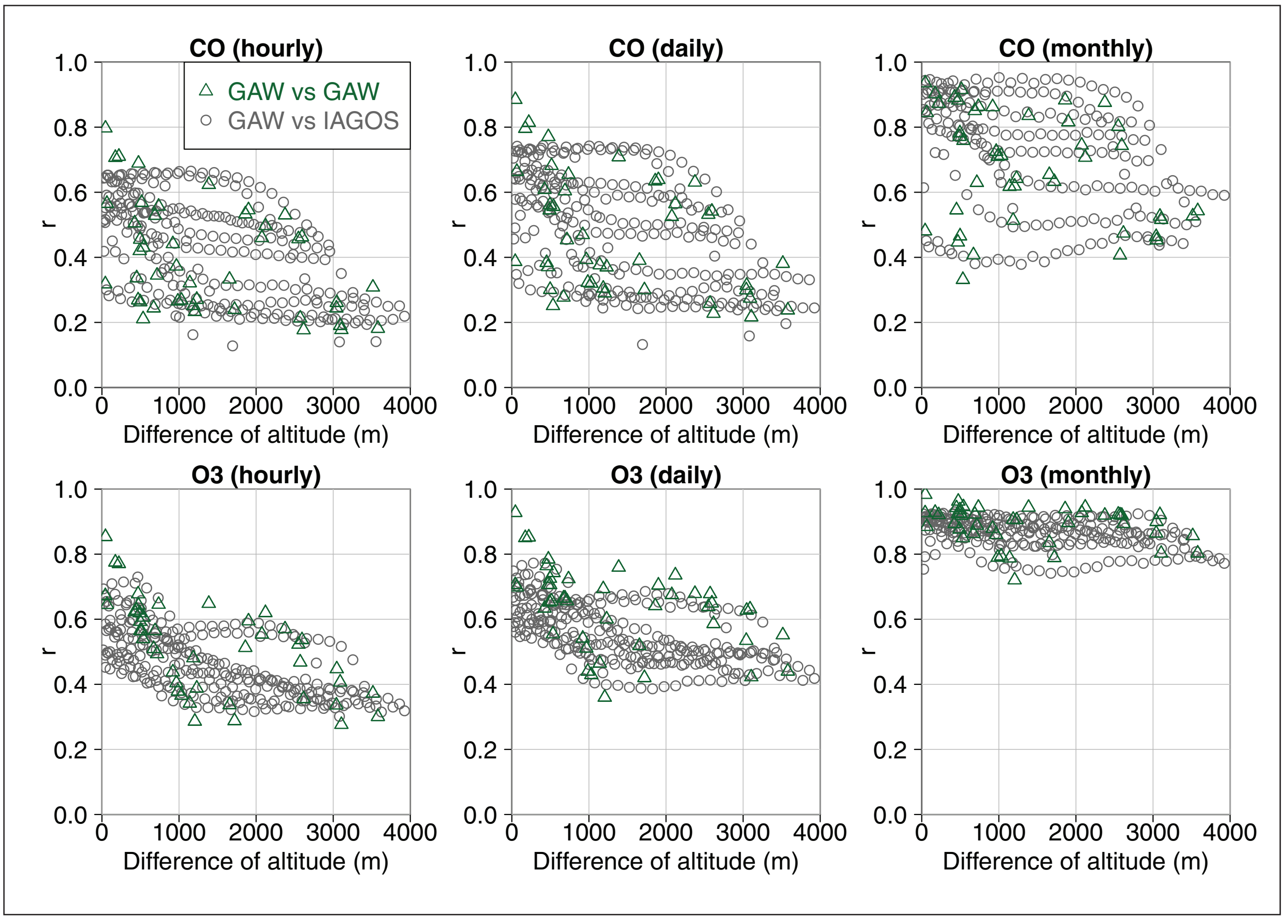

Figure 8: Correlation of CO and O_3 between IAGOS and GAW stations, and between GAW stations. This Figure shows the correlations between pairs of GAW stations (in green) and between GAW stations and IAGOS airborne observations (in grey), as a function of the difference of altitude (or elevation). Correlations are shown for hourly (left panels), daily (middle panels) and monthly (right panels) time series, for both CO (top panels) and O_3 (bottom panels). Each green triangle shows the correlation between two given GAW stations plotted against the difference of elevation between these two stations. Each grey circle represents the correlation between one given GAW station and the IAGOS data at one given altitude level, plotted against the difference of altitude between the two datasets (i.e. the difference between the elevation of the GAW station and the given IAGOS altitude level). For clarity, the correlations between GAW and IAGOS data are shown every 3 IAGOS altitude levels (i.e. at 25 m, 175 m, 325 m, etc. up to 4 km ASL). The correlations between IAGOS and GAW are found to be in the same order of magnitude of than the correlations among GAW stations themselves. Although variable, these correlations tend to be higher for lower differences of altitude. DOI: https://doi.org/10.1525/elementa.280.f8

3.7 Comparison between ascent and descent IAGOS profiles

In this section, we further discuss the potential influence of the airport pollution on the IAGOS measurements. One characteristic of the IAGOS data is that profiles can be obtained during either ascent (take-off) or descent (landing) phases. Although there are exceptions (see Figure S-7 in the Supplement for a plot of the aircraft orientation as a function of the wind direction for all concomitant flights at Frankfurt), the general rule is to take-off and land against the wind when possible, which helps slowing down the aircraft during the landing and increasing the lift on the wings during the take-off. Such operations are facilitated by the fact that runways are preferably built following the local dominant wind directions. For instance, the runways available at the Frankfurt airport offer four possible orientations for take-offs and landings: ENE, WSW, S, N (where N, S, E, W stand for North, South, East, West). Depending on the wind direction and the configuration of both the runways and the different airport facilities, and assuming an influence of the airport pollution on the IAGOS measurements, we might expect a difference of O_3 or CO mixing ratios during landing and take-offs. In particular, we might expect higher CO mixing ratios during the landing phase (when the wind blows the airport plume toward the aircraft) than during the take-off phase (when the aircraft get further from the airport with a headwind clear from any influence of airport emissions). Conversely, as O_3 is quickly titrated by the NO emissions, we might expect lower O_3 mixing ratios during the landing than during the take-off, although some O_3 local production may lead to a more complex and ambiguous picture.

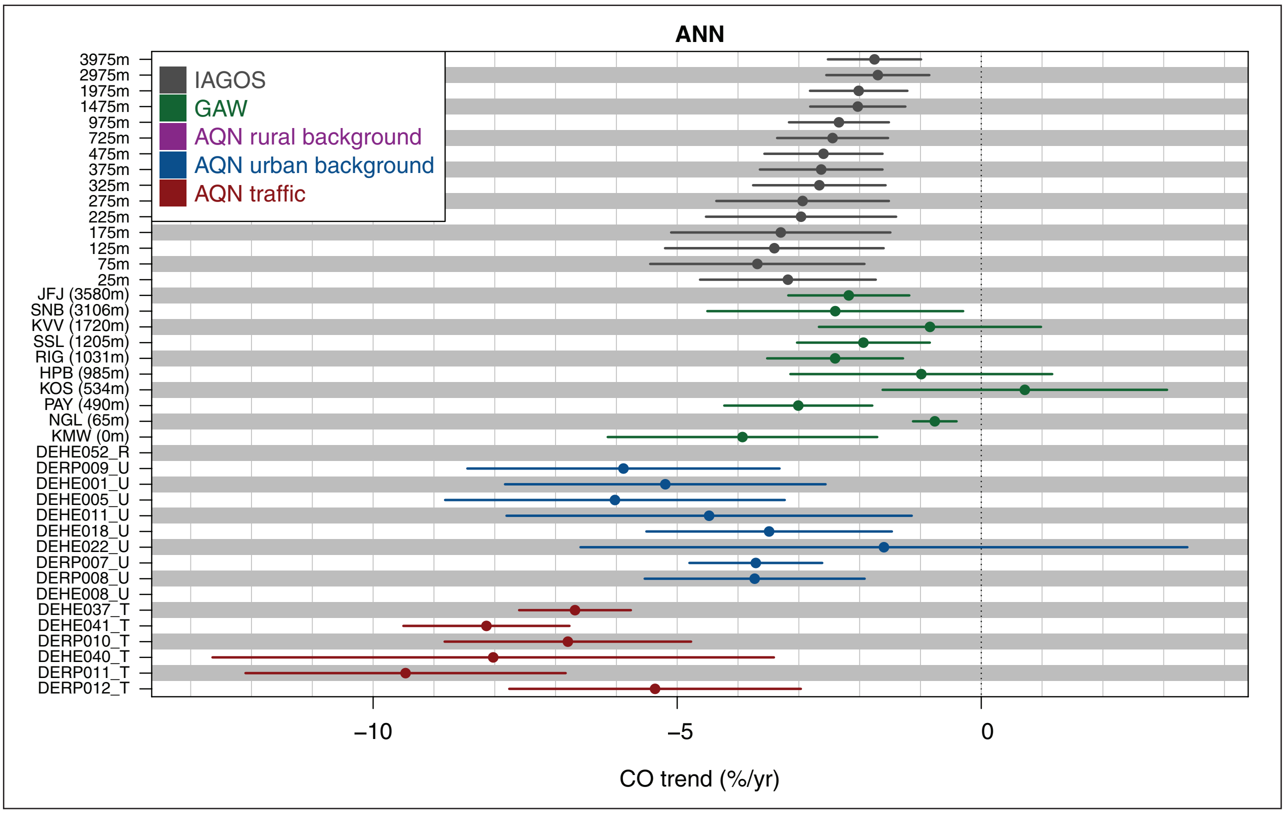

Figure 9: Relative annual trends of CO mixing ratios at Frankfurt, over the period 2002–2012. Trends are calculated by linear regression based on the time series of annual averages, with annual averages defined only when data are available during all four seasons. Uncertainties are shown at a 95% confidence level. The Figure highlights a strong decrease of CO at traffic stations (–5.5 to –10% yr^{-1}), reduced to about –2.5 to –6% yr^{-1} at urban background stations. At GAW stations, the significant trends are lower (from –4 to –1% yr^{-1}) and even not significant for several stations. Close to the surface, IAGOS data show significant negative trends with quite similar rates than urban background stations (about –4%). Higher in altitude, these negative trends decrease to rates typical of GAW stations (–2% at 4 km ASL). DOI: https://doi.org/10.1525/elementa.280.f9

To investigate this point, we compared the O_3 and CO IAGOS profiles during these two phases over the period 2002–2012. We selected at Frankfurt all concomitant landings and take-offs profiles over a time window of 30 min. The mean profiles are shown in **Figure 10**, with their corresponding uncertainties at a 95% confidence level (i.e. ±2 standard deviations normalized by the square root of the number of points). Contrary to what would have been expected if IAGOS data were more influenced by the airport pollution during landings, the CO mixing ratios are slightly higher during take-offs. Results show statistically insignificant biases of about 5% below 300 m ASL and (slightly) significant biases of about 10% between 300 and 900 m ASL, the strongest difference (15%) being found at 350–400 m ASL. Above 900 m ASL, the differences are much lower and remain insignificant. The agreement between landings and take-offs is very good for O_3 with a mean insignificant difference below 1% over the first kilometre.

If the airport emissions were influencing IAGOS data, one would expect higher CO mixing ratios during landing, especially close to the surface, which is not the case here. The differences previously highlighted may be at least partly explained by the spatial heterogeneity of the CO mixing ratios field in the boundary layer around Frankfurt. At low altitudes (below 300 m ASL), both the ascending and descending aircraft are closer to each other, leading to small (and insignificant) differences of CO mixing ratio. Higher in altitude between 300 and 900 m ASL, the horizontal distance between the two aircraft quickly increases (reaching typically 5–20 km) since they are often flying in the same direction (close to the wind direction). Considering the strongly heterogeneous spatial distribution of emission sources in the region of Frankfurt (patchwork of forests, urban and agricultural areas), this may explain a part of the differences between landings and take-offs. Note that take-off and landing observations are not similarly distributed around Frankfurt notably since the runway oriented S-N is only used for take-offs. If we go further in altitude (above 1 km ASL), the aircraft typically reaches the free troposphere where CO mixing ratio fields are more homogeneous. Conversely, the very low differences of O_3 found between landings and take-offs may be due to the fact, as a secondary pollutant, the O_3 fields are more homogeneous (relatively to CO).

Note that comparing daily or monthly averages based on all available data (rather than averaging only

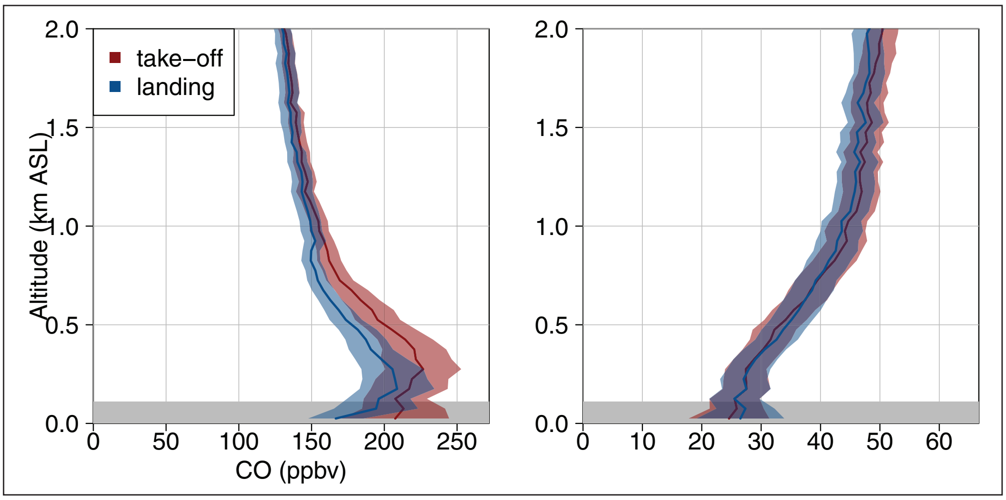

Figure 10: Mean vertical profiles of CO (left panel) and O_3 (right panel) for concomitant take-offs and landings over a time window of 30 min. The shaded area shows the uncertainty on the mean at a 95% confidence level (±2 standard deviations normalized by the square root of the number of points). The differences of CO mixing ratios are small (~5%) and statistically insignificant below 300 m ASL, and moderate (10–15%) and slightly significant between 300 and 900 m ASL. Above, the differences are low and insignificant. A very good agreement is found for O_3. DOI: https://doi.org/10.1525/elementa.280.f10

simultaneous landing and take-off data), stronger differences are found due to the fact that (i) both O_3 and CO depict a substantial diurnal variability in the lower troposphere, and (ii) landings and take-offs are not similarly distributed over the day in the IAGOS database (landings are more numerous during the morning while take-offs are more equally distributed along daytime).

3.8 Comparison with other airports in Europe

In the previous sections, we highlighted that the O_3 and CO mixing ratios typically measured by IAGOS aircraft in the first altitude levels at Frankfurt are close to the mixing ratio measured at urban background stations. Higher in altitude, they come closer to the mixing ratios observed at the regional scale by the GAW network. The progressive shift from a behaviour typical of urban background stations to a behaviour closer to the regional background was also highlighted on the temporal variability of O_3 and CO. These results are valid for Frankfurt, but the question arises of whether they can be generalized to other IAGOS airports. Compared with Frankfurt, IAGOS data are much sparser at other airports, which limits our ability to conduct similar in-depth analysis. However, some general comparisons can still be performed at few other airports in order to assess how far these previous results may change at other locations. In this section, we thus extend the previous analysis to two other airports of West-Central Europe: Vienna and Paris. The analysis is restricted to a comparison of the overall distribution of mixing ratios and a discussion on the correlation of hourly and daily datasets. For information purposes, the corresponding figures are given in the Supplement (Figures S-8 to S-10 for Vienna, Figures S-11 to S-13 for Paris). To facilitate comparisons among airports, the mean CO and O_3 mixing ratios at the different surface stations and in altitude are reported in **Table 2** for the 3 cities.

3.8.1 Vienna

The comparison between IAGOS and surface CO measurements in Vienna gives quite similar results as in Frankfurt. IAGOS data in Vienna are available only between 2002 and 2006. Considering only the days with available IAGOS data at one altitude level at least, the mean CO mixing ratio are lower than at Frankfurt, with 439 ± 268 and 272 ± 163 ppbv at traffic and urban background stations, respectively. Although it lies between these two values, the mean CO mixing ratio measured by IAGOS in the first altitude level (0–50 m ASL) is quite strong (347 ± 152 ppbv) compared to Frankfurt. However, this value is not fully representative as it is based on only 236 points (compared to more than 2,000 for altitude levels above 100 m) and more importantly, without any data during summertime (i.e. when CO is minimum). As mentioned in Sect. 2.1, this study is using the barometric altitude estimated from the temperature and the pressure measured by the aircraft assuming standard conditions at the surface. This can lead to barometric altitudes below the actual airport elevation (180 m for Vienna) under specific atmospheric conditions characterized by low pressure (cyclone) and/or low temperature. Such conditions are mostly encountered in winter and not in summer, which explains the absence of IAGOS data at this first altitude level in summer. In addition, these cyclonic conditions may be associated to stronger local CO emissions (for instance, a wintertime cold snap increases the need for residential heating). At the next levels, the CO mixing ratios measured by IAGOS at Vienna are in better agreement with results obtained at Frankfurt.

It is worth noting that exact differences between IAGOS and urban background stations cannot be calculated from the figures in **Table 2** as the mean urban background at the surface (here calculated from all hours with at least one IAGOS observation between 0 and 4 km ASL, see

Sect. 2.3) changes if we consider the IAGOS availability at one specific altitude level. Exact comparisons between all individual hourly surface data with concomitant IAGOS data at the corresponding altitudes (i.e. the elevation of the station) were performed, and results are reported in **Table 3** (results concerning Frankfurt have been already discussed in the previous sections). On average, the CO measured by IAGOS aircraft is 48 and 17% lower than at surface traffic and urban background stations, respectively, which remains in agreement with the results at Frankfurt (–61 and –26%, respectively).

For O_3, the mean mixing ratios are 33 ± 19 and 35 ± 19 ppbv at urban and rural background stations, respectively. Compared to Frankfurt, IAGOS data at Vienna show lower O_3 mixing ratios in the first levels, but the agreement is good higher in altitude (mean difference below 3 ppbv above 400 m ASL), in agreement with the results of Logan et al. (2012) in the low/middle troposphere (2.6–5 km). Considering exact comparisons between IAGOS and surface stations, the mean difference is only +2 and +10% for rural and urban background stations, respectively (**Table 3**). As for Frankfurt, these differences show diurnal variations with highest biases during the morning and lowest during the second half of the day (not shown). The correlation between the two datasets is strong (r = 0.82).

Therefore, for both CO and O_3, IAGOS aircraft at Vienna measure mixing ratios in agreement with the urban background observed at the surface by the close-by AQ monitoring stations. Despite a lower number of

Table 2: Mean CO and O_3 mixing ratios (ppbv) measured by rural background, urban background and traffic stations, and by IAGOS aircraft in the first 4 altitude levels, over the period 2002–2012. DOI: https://doi.org/10.1525/elementa.280.t2

Species	Airport	Rural background mean ± stdev (min–max[a])	Urban background mean ± stdev (min–max[a])	Traffic mean (min–max[a])	IAGOS			
					1st level	2nd level	3rd level	4th level
CO	Frankfurt	–	348 ± 212 (287–405)	630 ± 414 (467–792)	292 ± 142	261 ± 124	244 ± 109	230 ± 104
	Vienna	–	272 ± 163 (262–279)	439 ± 268 (323–623)	347 ± 152[b]	271 ± 126	222 ± 104	213 ± 96
	Paris	–	486 ± 264 (417–574)	1,322 ± 652 (813–1,782)	275 ± 168	242 ± 123	218 ± 93	207 ± 78
O_3	Frankfurt	35 ± 15 (35–35)	19 ± 17 (16–20)	–	17 ± 16	20 ± 17	20 ± 16	21 ± 16
	Vienna	35 ± 19 (33–36)	33 ± 19 (26–37)	–	21 ± 13[b]	26 ± 16	32 ± 19	35 ± 19
	Paris	28 ± 19 (26–29)	22 ± 19 (18–36)	–	19 ± 16	20 ± 16	21 ± 16	23 ± 16

[a] Minimum and maximum mean mixing ratios among the different surface stations.
[b] Low number of observations, and no data available during the summer (see text).

Table 3: Statistical results of comparisons between IAGOS and surface stations at the hourly scale. For a given type of station, species and airport, relative biases and correlations (r) are computed considering simultaneously all individual hourly data from all surface stations of that type against concomitant IAGOS data at the altitude level corresponding to the elevation of the stations. Is also indicated the number of hourly observations taken into account (N). NB: A positive bias means that IAGOS mixing ratios are higher than at surface stations. DOI: https://doi.org/10.1525/elementa.280.t3

Species	Airport	Rural background stations			Urban background stations			Traffic stations		
		Bias	r	N	Bias	r	N	Bias	r	N
CO	Frankfurt	–	–	0	–26%	0.58	28,976	–61%	0.40	24,559
	Vienna	–	–	0	–17%	0.80	1,384	–48%	0.64	1,833
	Paris	–	–	0	–48%	0.38	228	–82%	0.21	997
O_3	Frankfurt	+12%	0.85	6,608	+3%	0.81	32,541	–	–	0
	Vienna	+2%	0.88	1,437	+10%	0.84	7,605	–	–	0
	Paris	–23%	0.76	6,095	–12%	0.81	9,242	–	–	0

observations available, the correlation profiles of CO at Vienna (Figure S-10 in the Supplement) are also in good agreement with those obtained at Frankfurt. For O_3, quite similar correlations are found in both cities, but some differences are found in the shape of profiles. Strongest close to the surface, the correlations between IAGOS and AQN stations are found to decrease more slowly and more uniformly over the first 4 kilometres (while the decrease of correlation at Frankfurt was occurring mainly in the first kilometre). They usually remain similar or slightly higher than the correlations between IAGOS and GAW stations.

3.8.2 Paris

At Paris, we combine the measurements of both Charles-de-Gaulle and Orly airports (most data are obtained at the former airport). The observed pollution from CO is much stronger than in the other two cities, with 486 ± 264 ppbv at urban background stations and 1,322 ± 652 ppbv at traffic stations on average. These numbers are biased high as they are calculated based on a limited number of points with concomitant IAGOS observations, all during the beginning of the period (2002–2005). Independently of IAGOS, the negative trends in CO observed between 2002 and 2012 at the urban background stations in Paris (not shown) are higher than at the two other airports (about –7% yr^{-1}, against less than –5% yr^{-1} at other airports), leading to a mean CO over the period 2002–2012 reduced to 294 ± 209 ppbv. This is also the case for traffic stations (trends ranging from –8 to –12% yr^{-1}, against less than –8% yr^{-1} at other airports) except that the mean CO over 2002–2012 is still higher than at the other airports (881 ± 585 ppbv).

The mixing ratios measured at Paris by IAGOS aircraft are much lower than at urban background stations. The mean urban background of CO is calculated based on only two stations, including one in the centre of Paris (PA1H_U: Paris 1st district) and the another one (AUB_U: Aubervilliers) in the adjacent suburbs (350 m from the Paris ring road highway). On average, the relative difference between IAGOS and these urban background stations is –48% (**Table 3**), thus much stronger than at Frankfurt or Vienna. Although the number of data points is a lot lower than at the two other airports (228 data, against 1,384 and 28,976 for Vienna and Frankfurt, respectively), the difference is statistically significant. Based on more numerous data, the difference between IAGOS and traffic stations remains higher than at other airports (–82% on average). For both types of station, the correlation is also much lower (0.38 and 0.21, respectively). The poor agreement can be at least partly explained by the IAGOS data availability predominantly in the morning hours (55% of the 236 points are between 07:00–10:00 LT, 74% between 06:00–11:00 LT). As mentioned in Sect. 3.2, the difference between IAGOS observations and surface observations at urban background stations varies diurnally, with larger biases during the morning rush hours. A similar behaviour is observed at Paris (not shown), although IAGOS CO mixing ratios remain substantially lower than at urban background stations.

Compared with CO, there are many more data from stations measuring O_3 in/around Paris (8 rural background stations, 21 urban background stations) available. They show mean O_3 mixing ratios of 28 ± 19 and 22 ± 19 ppbv at the rural and urban background stations, respectively. The IAGOS data show slightly lower O_3 mixing ratios, with mean differences of –22 and –11% for rural and urban background stations (**Table 3**). Similarly to Frankfurt and Vienna, both are well correlated ($r > 0.7$).

The bias of O_3 between IAGOS and rural background stations can be greatly reduced by restricting the dataset to days with windy conditions. This is clearly illustrated in **Figure 11** in which the relative bias is shown for different values of minimum wind speed from 0 to 10 m s^{-1}. This bias progressively decreases when data associated with low wind speed (measured by IAGOS aircraft) are removed: applying a minimum wind speed of 2, 4, 6, 8 and 10 m s^{-1} leads to a bias of –21, –16, –14, –11 and –10%, respectively. For wind criteria above 8 m s^{-1}, biases are not significant. However, such restrictions of the dataset slightly reduce the correlations. Such an influence of the wind was not observed at the other airports. A good agreement between IAGOS and rural background stations is not expected under very low wind conditions, as mixing in the vicinity of the surface station is reduced. However, the persistent decrease of the bias for higher wind speed was not expected. It may be due to the flat orography of the Paris region that favours homogenisation of the spatial O_3 distribution at a regional scale when the wind is sufficiently high (contrary to Frankfurt and Vienna where the surrounding orography is more complex). Note also that a similar reduction of the bias with the wind speed is highlighted between IAGOS and the EIFF3_U station located at the 3rd floor of the Eiffel tower (315 m ASL).

4 Discussion and conclusion

The CO and O_3 mixing ratios measured by IAGOS at different altitude levels in the lower troposphere around three European airports have been compared with surface measurements available from local air quality monitoring networks (within 50–80 km from the airport) and regional GAW stations (within 500 km from the airport). A focus was made on Frankfurt airport where the IAGOS data record is the densest and longest. At Frankfurt, the measurements at (urban) traffic stations show a very specific behaviour, with very high CO mixing ratios, and a strong negative trend over the period 2002–2012. Conversely, the CO and O_3 measured at urban background stations were found to share common characteristics with the CO and O_3 measured by IAGOS in the altitude levels closest to the surface, in terms of distribution, seasonal variations and trends (**Figures 3, 6, 9**). IAGOS data showed slightly lower CO and slightly higher O_3, which suggests a smaller influence of local emission sources than at urban background stations. These differences exhibit a diurnal variation with largest biases during the morning rush hours when the local traffic emissions increase the CO mixing ratios at both urban background and traffic stations (**Figure 4**). In addition, the CO and O_3 mixing

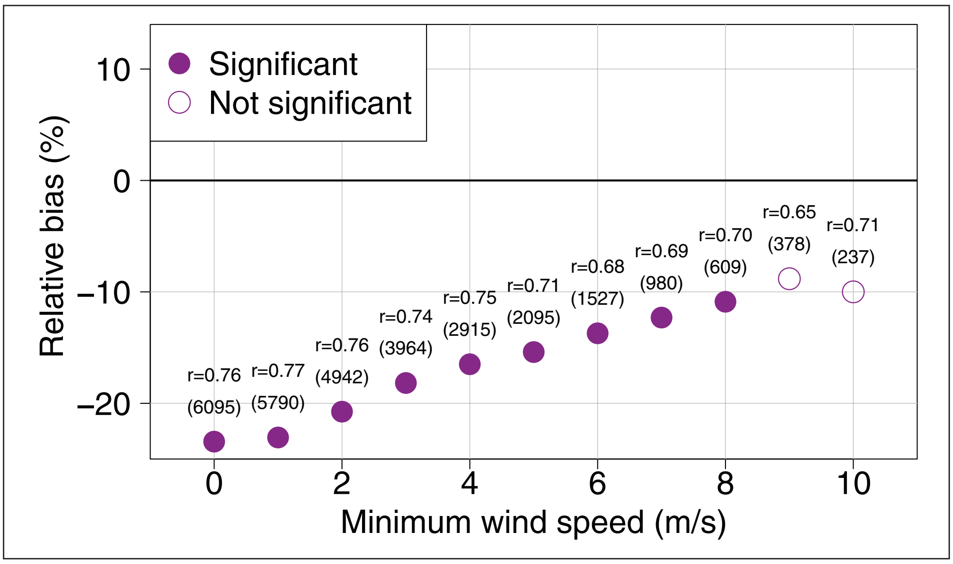

Figure 11: Influence of the wind speed on the O_3 bias between IAGOS and rural background stations at Paris. The Figure shows the relative bias of O_3 between IAGOS and rural background stations depending on the minimum wind speed. A total of 8 rural background stations are included. The wind speed is measured by IAGOS aircraft. The bias is calculated taking into account simultaneously all hourly data from all stations and concomitant IAGOS data at the corresponding altitude levels. The filled circles indicate a statistically significant difference (at a 95% confidence level) while the empty circles indicate that the difference is not significant (too low number of points and/or too small difference). The correlation and the number of hourly values taken into account (in bracket) are also reported. The Figure clearly highlights a growing agreement between IAGOS and rural background stations as the wind speed increases. DOI: https://doi.org/10.1525/elementa.280.f11

ratios measured at the nearby surface stations appeared reasonably well correlated with the IAGOS airborne data at the lowest altitude levels (r ~ 0.6–0.7 at the daily scale, 0.7–0.8 at the monthly scale) (**Figure 7**). At higher altitudes, these characteristics of the IAGOS observations quickly deviate from the urban background and approach the regional background observed by the different GAW surface stations located in and around Germany. This is demonstrated by the fact that the correlation between IAGOS and GAW datasets increases with altitude, with maximum correlations mostly obtained at an altitude close to the elevation of the surface station (**Figure 6**). The correlation between IAGOS and GAW stations is in the same range as the correlation among the GAW stations themselves (**Figure 8**). The vertical distribution of CO and O_3 derived from the IAGOS airborne measurements was found to be in close agreement with the vertical distribution deduced in the first 4 km from the GAW stations at different elevations (**Figure 5**). A comparison between concomitant ascent and descent profiles at Frankfurt have shown a good agreement (insignificant bias) on CO below 300 m ASL and above 1,000 m ASL but some small significant differences between 300 and 1,000 m ASL, with lower CO during landings (**Figure 10**). The agreement on O_3 was very good whatever the altitude. Despite sparser IAGOS data, some comparisons were extended to Vienna and Paris airports. In both cities, the CO mixing ratios measured by IAGOS in the first altitude levels are always lower than the urban background given measured at the surface. At Vienna, the agreement with Frankfurt is good, as illustrated by a mean difference of CO between IAGOS and urban background stations of –17% on average (against –26% at Frankfurt). At Paris, much lower CO mixing ratios are measured by IAGOS (difference of –46%), at least partly due to a high proportion of data during the morning, when higher biases were also observed at Frankfurt and Vienna. With biases below ±12%, the agreement of O_3 between IAGOS and urban background stations appears very satisfactory at all 3 airports. Although some quantitative differences may exist, the results at Vienna and Paris remain reasonably consistent with the results obtained at Frankfurt.

Therefore, several important conclusions can be drawn at these airports. Firstly, the IAGOS observations close to the surface do not appear to be strongly influenced by local emissions from either the airport activities on the tarmac or the other aircraft sharing nearby flight tracks. Secondly, the comparison with the surrounding surface stations from the local AQN indicates that the IAGOS observations in the first few hundreds meters above the surface have a representativeness typical of urban or more precisely suburban background stations where the three airports are located. Thirdly, the comparison with (more distant) regional background stations from the GAW network shows that as one moves higher in altitude, the IAGOS observations shift toward a regional representativeness. The boundary of transition between these two variability regimes likely depends on the local meteorological conditions, and in particular the PBL height and state. The transition may thus be more abrupt than our results suggest since in this study, numerous profiles of different PBL height are combined, which automatically smoothes the features that may exist at the transition between PBL and lower free troposphere. The good consistency between GAW and IAGOS observations at the different altitudes (more particularly above the first 500 m) gives confidence on the ability of IAGOS data to measure a reliable vertical distribution of the pollutants in the lower troposphere.

Several reasons can explain the absence of strong influence of the local emissions from the airport activities and/or other aircraft. First, although CO mixing ratios as high as a few ppmv can typically be reported in international airports (e.g. Yu et al., 2004; Schürmann et al., 2007), the IAGOS system is designed to start measurements when the wheels of the aircraft leave the ground during ascents (or when they touch down during descents). At this moment, the aircraft is already moving at a high speed (typically 50 m s^{-1}) and is thus expected to quickly get away from the local emissions associated with the airport activities. A standard landing and take-off (LTO) cycle as defined by the International Civil Aviation Organization (ICAO) includes 4 phases with different durations and percentages of thrust: the approach phase (30% thrust, 4 min), the idle(-taxi) phase (7% thrust, 26 min), the take-off phase (100%, 0.7 min) and the climb phase (85% thrust, 2.2 min) (Masiol and Harrison, 2014). IAGOS measurements thus start at the beginning of the climb phase for ascents, and at the end of the approach phase for descents. The emission factors of CO are much higher during the idle phase than during the other phases (see Masiol and Harrison, 2014 for a review). This is due to a less complete combustion associated with cold combustor temperatures in the aircraft engines (Herndon et al., 2008). However, IAGOS is not measuring during this phase, which likely explains the absence (or the very low frequency of occurrence) of strong CO peaks: indeed, between 0 and 250 m ASL at Frankfurt, the maximum CO mixing ratio is 2,358 ppbv, and the 99.9th percentile of all hourly mixing ratios is only 1,095 ppbv. Secondly, in order to reduce the risk of collision and/or the exposure to the wake vortex turbulence caused by other aircraft, several separation standards are fixed by the ICAO. For instance, the vertical separation minimum between two aircraft is fixed to 300 m (1,000 ft) below ~8.8 km of altitude (and the double above) (ICAO, 2016). In addition, although the minimum duration between two consecutive takeoffs/landings can be as short as a few minutes (it varies depending on the aircraft and the airport), commercial aircraft are not allowed to follow each other too closely on the same track. Considering the fact that the dispersion of pollutants is more efficient at altitude (stronger wind, absence of re-circulation loops caused by buildings and/or canyon streets), these different separation standards appear sufficient to avoid measuring directly the exhausts from other aircraft. Detailed studies on airport air quality aspects with IAGOS data are subject of on-going work.

Therefore, this study shows that IAGOS can be seen as a complement of surface stations to monitor the air quality in/around the agglomeration. Although sporadic in time, IAGOS data provide useful information on the vertical distribution of pollution in the boundary layer. The spatial representativeness of the IAGOS observations in the lower troposphere appears large enough to offer a new opportunity to validate models and satellites in the PBL, in complement to surface stations and ozonesondes. The importance of improving models on the vertical dimension has been already underlined in several studies (Solazzo et al., 2013; and references therein). Besides, this has recently justified the use of IAGOS to validate operationally the CAMS regional chemistry-transport models in Europe (see http://www.iagos.fr/cams for daily comparisons). This study confirms that such comparisons are entirely relevant. In addition, IAGOS offers the major advantage of providing observations in many large cities with the same instrumental system, thus ensuring a consistency between all the measurements. This is particularly interesting for comparing the air quality in cities from different countries that may not use comparable instruments and standard scales at the surface stations.

It is worth remembering that these results are based on a limited set of airports. In particular, the most in-depth analysis was restricted to Frankfurt airport where the IAGOS data are the most numerous. Considering the potentially large differences of environment from one airport to the other (e.g. intensity of the local air traffic, distance from the agglomeration, size of the agglomeration and intensity of local emissions, orography, local meteorology), they may not be valid for all airports visited by IAGOS aircraft since 1994 (~290). This study thus has to be considered as a first step, and should be extended in the future as larger amounts of data become available around the different airports, in particular in highly polluted Asian megacities.

Data Accessibility Statement

No new measurements were made for this review article. All datasets mentioned in the text were obtained from existing databases. The IAGOS data are available on: http://www.iagos.fr, the AIRBASE data (for Frankfurt and Vienna) on: https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-7, the AIRPARIF data (for Paris) on: https://www.airparif.asso.fr. The GAW data are taken from the World Data Centre for Greenhouse Gases (WDCGG) http://ds.data.jma.go.jp/gmd/wdcgg/.

Supplemental Files

The supplemental files for this article can be found as follows:

- **Figure S-1.** Location of the IAGOS airborne observations at the Frankfurt airport. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-2.** Location of the IAGOS airborne observations at the Vienna airport. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-3.** Location of the IAGOS airborne observations at Paris airports. DOI: https://doi.org/10.1525/elementa.280.s1

- **Figure S-4.** Monthly time series of CO mixing ratios at Frankfurt. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-5.** Monthly time series of O_3 mixing ratios at Frankfurt. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-6.** Relative annual trends of O_3 mixing ratios at Frankfurt, over the period 2002–2012. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-7.** Orientation of the aircraft against the wind direction at Frankfurt. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-8.** Distribution of the hourly CO (top panel) and O_3 (bottom panel) mixing ratios at the GAW (Global Atmospheric Watch) and AQN (Air Quality Network) surface stations and at different IAGOS altitude levels at Vienna. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-9.** CO (top panel) and O_3 (bottom panel) mixing ratios versus altitude, as given by IAGOS and surface stations at Vienna. The grey area indicates the elevation of the Vienna (180 m) airport. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-10.** Correlation between the IAGOS observations at several altitudes and the surface measurements, at Vienna. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-11.** Distribution of the CO (top panel) and O_3 (bottom panel) hourly mixing ratios at Paris. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-12.** CO (top panel) and O_3 (bottom panel) mixing ratios versus altitude, as given by IAGOS and surface stations at Paris. DOI: https://doi.org/10.1525/elementa.280.s1
- **Figure S-13.** Correlation between the IAGOS observations at several altitudes and the surface 3 measurements, at Paris. DOI: https://doi.org/10.1525/elementa.280.s1
- **Table S-1.** Description of the surface stations. DOI: https://doi.org/10.1525/elementa.280.s1

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Competing interests

The authors have no competing interests to declare.

Author contributions

- Contributed to conception and design: HP, MJ, VT, BS
- Contributed to acquisition of data: HP, VT, BS, GA, RB, DB, J-MC, PN, MS
- Contributed to analysis and interpretation of data: HP, MJ, VT, BS, FG, HC, MS
- Drafted and/or revised the article: HP
- Approved the submitted version for publication: HP, MJ, VT, BS, GA, RB, DB, J-MC, PN, FG, HC, MS

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