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A new method for estimating emission ratios in the urban atmosphere: examples of ratios to CO₂, CO and volatile organic compounds in Paris

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Abstract

We propose a new approach to estimate urban emission ratios that takes advantage of the enhanced local urban signal in the atmosphere at low wind speed. We apply it to estimate monthly ratios between CO₂, CO and some VOCs from several atmospheric concentration measurement datasets acquired in the centre of Paris between 2010 and 2014. We find that this approach is little sensitive to the regional background level definition and that, in the case of Paris, it samples all days (weekdays and weekends) and all hours of the day evenly. A large seasonal variability of the $\Delta\text{CO}/\Delta\text{CO}_2$ ratio in Paris is shown, with a difference of around 60 % between the extreme values and a strong anti-correlation ($r^2 = 0.75$) with atmospheric temperature. The comparison of the ratios obtained for two short measurement campaigns conducted in two different districts and two different periods (fall and winter) shows differences ranging from -120 to +63 %. A comparison with a highly resolved regional emission inventory suggests some spatial variations of the ratio within the city, although most of these differences seem to be rather driven by the seasonal variability.

1 Introduction

In response to changing air quality and climate, there is a growing interest in quantifying emissions of pollutants and greenhouse gases from urban areas (UNEP, 2013; EEA, 2014). Urban emissions are usually known through the combination of direct and indirect geospatial energy use statistics with emission factors for individual source sectors. The heterogeneity of the input data in space, time and type makes it difficult to monitor the uncertainties of these inventories. Such monitoring actually receives little incentive at the international level (e.g., Bellassen and Stephan, 2015), but it has been an active topic for scientific research. Some studies have been based on measurement campaigns dedicated to specific sectors, for instance air-composition measurements in road tunnels for traffic emissions (e.g., Touaty and Bonsang, 2000; Ammoura et al.,

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(Association in charge of monitoring the air quality in the Paris region) provided for year 2010, emissions of pollutants and greenhouse gases are mainly from the traffic (29 %) and residential and service sectors (43 %) (Airparif, 2013).

The paper is structured as follows. Section 2 presents the measurements and the data. Section 3 starts with a presentation of typical measurements and a discussion about the choice of the background level, presenting two different options. The analysis method itself to estimate urban emission ratios is described in Sect. 3.3 including sensitivity tests (Sects. 3.3.2 and 3.3.3). Section 4 presents the results obtained for different periods of the year and different years. Section 4.1 gives the interpretation of the ratios determined with our method and discusses the representativeness of these ratios. Section 4.2 presents the seasonal variability of the $\Delta\text{CO}/\Delta\text{CO}_2$ ratio in Paris and Sect. 4.3 compares all ratios between co-emitted species obtained during two short campaigns in Paris.

2 Methods

2.1 Site description

All atmospheric composition measurements presented in this study have been made in the centre of Paris. The instruments were installed at two sites. The first one is located on the Jussieu campus of University Pierre et Marie Curie (UPMC) at the QualAir station (<http://qualair.aero.jussieu.fr>). This station stands on the roof of a building, on the left bank of the river Seine (48°50' N, 2°21' E and 23 m a.g.l.). A botanical garden of 28 ha, the Jardin des Plantes, lies about 500 m from the measurement site. The closest motorways are about 4 km on the south and on the south-east, but the university is surrounded by many streets which are particularly congested during rush hours. The emission activities in the centre of Paris essentially originate from road traffic activities and from the residential and service sectors, since most industrial activities have been removed in the 1960s (AIRPARIF, 2013).

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The second measurement site is the roof of Laboratoire d'Hygiène de la Ville de Paris (LHVP) located about 2 km from the Jussieu campus, south-east of it (48°49' N and 2°21' E and 15 m a.g.l.). It dominates a public garden of 4.3 ha, the Parc de Choisy. Residential buildings and arterial roads also surround this site. The closest motorway is a few hundred meters south of the site.

2.2 Instrumentation and air sampling

2.2.1 Joined MEGAPOLI/CO₂-Megapolis winter campaign

Our first campaign was performed jointly within the MEGAPOLI European project (Megacities: Emissions, urban, regional and Global Atmospheric POLLution and climate effects, and Integrated tools for assessment and mitigation project, <http://megapoli.info/>) and the CO₂-Megapolis project (<https://co2-megapolis.lsce.ipsl.fr>). This “winter campaign” took place in Paris during January–February 2010 (Dolgorouky et al., 2012; Lopez et al., 2013).

Two instruments were deployed at the LHVP. A Gas Chromatograph equipped with a Flame Ionisation Detector (GC-FID, Chromatotec) sampled Non-Methane Hydrocarbons (NMHCs). Mole fractions of alkanes, alkenes, alkynes and aromatic compounds were obtained with a time resolution of 30 min (air is sampled during the first 10 min and analysed during the next 20 min). More details can be found in Gros et al. (2011) and Dolgorouky et al. (2012).

A Cavity Ring-Down Spectrometer (CRDS G1302, Picarro Inc.) was also deployed to analyse CO₂, CO and H₂O mole fractions with a time resolution of 1 s (see Lopez et al., 2013, for more details).

2.2.2 Long-term continuous CO₂ and CO measurements

A Cavity ring-Down analyser (CRDS G1302, Picarro Inc.) performed continuous CO₂, CO and H₂O measurements in Jussieu continuously from 4 February 2013 to 11

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time stamp corresponding to the beginning of the measurement. Thus for each compound measured by the other instruments (CRDS and meteorological instruments), data have been averaged on the same 10 min interval. Finally, in this study, all the data have a same time step of 30 min.

3 Results

3.1 Typical time series and identification of specific meteorological events

Fig. 1 shows an example of atmospheric gas dry air mole fractions time series collected during the Multi-CO₂ campaign in 2013, with a time step of 30 min. The wind speed during the same period is also represented on Fig. 1e. Time series recorded during the joined MEGAPOLI/CO₂-Megaparis campaign in 2010, as well as the continuous measurements of CO₂ and CO in Jussieu are shown in the Supplement.

Mole fractions of the different species appear to co-vary much, despite the different lifetime of the species: CO₂ and CO have typical life time in the atmosphere (τ) much longer than the observation period whereas acetylene has a τ of a 13 days and ethylene has a τ of a few hours. In comparison, the meteorological events in Paris during the campaign lasted from a few hours to one day so that VOCs with a τ longer than two days, like acetylene, can be almost considered as non-reactive species. For shorter-lived species, here only ethylene and propene (1 day $> \tau > 5$ h), we computed the correlations between these species and acetylene. When considering all the data of the Multi-CO₂ campaign (without any selection), coefficients of determination are high ($r^2 > 0.70$). These tight correlations between VOCs with different reactivity suggest a limited impact of the chemistry.

In Fig. 1, we identify some events when the mole fractions of all species were significantly higher than elsewhere over the campaign duration (1.25 to 6 times as high). These periods (30 and 31 October, 10 and 11 November) appear to be systematically linked to specific meteorological conditions when the wind speed was very low (less

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et al., 2011), but are particularly expensive to make and are not available for our study period. For short-lived species, the definition of the background is not as critical and the smallest measured value is often used.

Here, we investigate two options to define the urban background levels. The first option takes advantage of the fact that the urban emissions are positive fluxes, i.e. which increase local atmospheric mole fractions. We define background mole fractions as all measurements smaller than the fifth percentile of the species over a moving window. The moving window allows accounting for the dependence of the background on the synoptic situation or on the time of year, as the background changes seasonally for many gases. As the average characteristic time of synoptic changes is a few days, and in order to gather a significant amount of data, we define overlapping windows of three days that start every day at 00:00 UTC, in increments of 1 day. Fig. 1 displays the selected lowest 5 % as black disks for some species measured during the Multi-CO₂ campaign. In order to avoid discontinuities, we linearly interpolate the selected data to obtain a background mole fraction time series with a time resolution of 30 min (black curves on Fig. 1).

This background definition is simple to implement because it does not require additional measurements. It is expected to work well for all species that do not have local sinks in the atmosphere or at the surface. We saw in Sect. 3.1 that chemical sinks could be neglected for our measurements, but in the case of CO₂ during the vegetation-uptake season (summer in particular), vegetation within Paris also contributes to populating the fifth percentile.

Our second option (for CO₂ only) defines the background from a publicly available analysis of the global atmospheric composition. We test it for CO₂, the species for which the first definition may be the least appropriate. The definition of the background level of CO₂ relies on the global inversion product of the Monitoring Atmospheric Composition and Climate project (MACC v13.1, <http://www.copernicus-atmosphere.eu/>, Chevallier et al., 2010). This product has a resolution of 3.75° × 1.9° (longitude–latitude) in space and of 3 h in time. It combines the information from 131 CO₂ stations over the

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globe and a transport model within a Bayesian framework and estimates the CO₂ surface fluxes over the globe together with the full 4-D CO₂ field.

We extracted the 3 hourly time series of the CO₂ concentrations from the MACC database for the eight grid points that surround our two measurement sites, Jussieu and the LHVP. The CO₂ background mole fraction is estimated as the linear interpolation in time of the analysed CO₂ concentrations averaged over the eight grid points. In the following, we call $\Delta\text{species}$, the mole fractions excess from the background as defined by either method.

3.3 Determination of the ratios between co-emitted species

3.3.1 Description of the method

We present next the method to evaluate ratios of excess mole fractions between 2 species ($\Delta\text{species}_1$ and $\Delta\text{species}_2$). We consider a moving window of 4 h in increments of 30 min (each period contains 8 points). On each period, we compute the coefficient of determination r^2 between $\Delta\text{species}_1$ and $\Delta\text{species}_2$ and use a linear regression to evaluate the slope. This slope defines a ratio between the two considered $\Delta\text{species}$ over the 4 h period. We also calculate the difference between maximum and minimum $\Delta\text{species}_1$, which is plotted on the x axis, over this period (we name it $\delta\Delta\text{species}_1$). The motivation for this amplitude computation will be developed in Sect. 4.1. These calculations are made if more than 5 points exist during the time period and if species excesses are linearly related (a p value test relative to linear relationship of species excesses is conducted and p value < 0.001 are selected). As an example, on a 4 h period, we compute (i) the coefficient of determination r^2 between ΔCO and ΔCO_2 , (ii) the slope, which well fits the considered dataset (thus giving the $\Delta\text{CO}/\Delta\text{CO}_2$ ratio over this period) and (iii) $\delta\Delta\text{CO}_2$.

In Fig. 3, we show some examples of ratios determined on each 4 h period against the local corresponding species offset $\delta\Delta\text{CO}_2$. They have a simple structure with a horizontal asymptote when $\delta\Delta\text{CO}_2$ is high. The equation of the asymptote defines

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ratios and differences between derived ratios do not exceed 10%. The data selection for several ratios, including $\Delta\text{CO}/\Delta\text{CO}_2$, is presented on Fig. 3.

3.3.3 Sensitivity to the background choice

In this section, we test the influence of the chosen background definition on the obtained $\Delta\text{CO}/\Delta\text{CO}_2$ ratio using the methods described in Sect. 3.3.1. We compare $\Delta\text{CO}/\Delta\text{CO}_2$ ratios for 2013 using the 5th percentile or MACC simulations as background levels (MACC simulations for 2014 were not available when this study was conducted). The evolution of the ratios for both options is presented in Fig. 5. We evaluate the relative difference between the ratios derived from the two options (in % of the ratio obtained with the fifth percentile as background). Differences vary from -17% in August to +11% in September 2013. The highest differences are found for the summer months (11% on average), and the lowest ones for the winter months (3.2% on average). These results show that the definition of the background does not significantly affect the derived ratios, even during the summer months. This comes from the fact that urban mole fractions during low wind speed periods are usually enough larger than the background mole fractions (from around 1.25 to 6 times more).

After these analyses, we finally choose to define background levels using the fifth percentile on a running window of 3 days as described in Sect. 3.2. However, tests were conducted using the tenth percentile (and a running window of 3 days) or changing the length of the running window between 1 and 5 days (but still considering the fifth percentile). No significant difference was found using the tenth percentile (less than 2% difference between the two derived $\Delta\text{CO}/\Delta\text{CO}_2$ ratios). Comparing $\Delta\text{CO}/\Delta\text{CO}_2$ ratios obtained with different lengths of the running window, ratios differ by less than 6% from one case to another, thus consolidating our choice for background levels.

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4 Discussion

We apply the method presented in Sect. 3.3.1 to assess ratios between co-emitted species in Paris. In this section, we first discuss the interpretation and the representativeness of the ratios determined using the method previously presented. Then, we divide the analysis in two parts. First we focus on the seasonal variability of the $\Delta\text{CO}/\Delta\text{CO}_2$ ratio using continuous measurements acquired from February 2013 to June 2014. Then we compare the ratios between co-emitted species and CO_2 obtained for the two short campaigns (in Sect. 4.3).

4.1 Interpretation and representativeness of the ratios determined with the asymptotic method

The x axis in Fig. 3 ($\delta\Delta\text{species}_1$) represents the variability of the species excess over a 4 h period. Large values correspond to a strong increase or decrease in the species local emissions, and highlight the concentration peaks that occur at low wind speed. The presence of an asymptotic value in the monthly ratio plots like that of Fig. 3 suggests that the ratios do not vary much within the month. This stability is also confirmed by the regular spread of the selected events throughout the month. For instance, applying our method to the continuous CO and CO_2 measurements acquired in 2013/2014 in Paris, we notice that all days (weekdays and weekends) and all hours of the day were sampled equally: no period type is systematically missing (see Fig. 4). This feature allows our method to yield a robust average ratio per month in Paris.

4.2 Seasonal variability of the $\Delta\text{CO}/\Delta\text{CO}_2$ ratio in Paris

The evolution of the $\Delta\text{CO}/\Delta\text{CO}_2$ ratios in Jussieu between March 2013 and May 2014 is presented in Fig. 5. It shows a large seasonal variability with a maximum value in winter and a minimum value in summer. There is a difference of around 60%

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can occur for vehicles or heating systems). Secondly, these differences may highlight the importance of the seasonal variability of the ratios, which was shown in Sect. 4.2. Indeed, measurements were performed in autumn (October–November) for the Multi-CO₂ campaign and in winter (January–February) for the MEGAPOLI/CO₂-Megapolis one. The $\Delta\text{CO}/\Delta\text{CO}_2$ ratio from the latter campaign is also reported in Fig. 5 for the corresponding month of the year: it aligns well on the seasonal variability observed in Jussieu, even though this campaign was made four years before. Furthermore, average temperatures during the low wind speed periods were not the same: 10 °C during the Multi-CO₂ campaign, 3 °C during the MEGAPOLI/CO₂-Megapolis one. This is in agreement with the argument developed in Sect. 4.2: residential heating is more important in the heart of winter and its emissions make the $\Delta\text{CO} / \Delta\text{CO}_2$ ratio higher. Finally the instruments were not installed at the same location in the centre of Paris (there are 2 km between the two locations). Thus the emission area of influence could be different because the local activities are not exactly the same around the two sites. As an example, highways, where the vehicle speed is limited to 80 km h⁻¹ and the vehicle flow is high, are closer to the LHVP (MEGAPOLI/CO₂-Megapolis measurements), leading this site to be more influenced by large traffic emissions. This spatial variability of the ratios in Paris is confirmed by the latest Paris emission inventory Airparif 2010. Airparif provides annual CO and CO₂ emissions by districts in Paris. Jussieu is in the 5th district and the LHVP in the 13th. According to the latest Airparif inventory, the annual CO/CO₂ ratios are respectively 2.43 and 3.74 ppb ppm⁻¹ for the 5th and the 13th districts. However, the good agreement between the ratio from the MEGAPOLI/CO₂-Megapolis campaign (measurements in 2010) and the one derived in Jussieu (measurements in 2014) indicates that the seasonal variability is the main driver for the evolution of the ratios.

5 Conclusions

We have investigated the possibility to characterise local urban emissions through atmospheric mole fraction measurements collected during low wind speed periods. In the

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case of Paris, we have shown that this approach significantly reduces the sensitivity of the results to the species background level definition, even in the case of CO₂. Thanks to long-term continuous measurements, we have also shown that the low wind speed conditions in the centre of Paris (especially in Jussieu) sample the hours of the day and the days of the week rather evenly, so that the method characterises an average urban atmosphere.

The comparison of ratios obtained for the two measurement campaigns, Multi-CO₂ and MEGAPOLI/CO₂-Megaparis, shows differences from –120 to +63 % for 9 atmospheric species. Such differences may reveal spatial and seasonal variability in the ratios because the two campaigns took place at different sites, during different years and seasons. However, the evolution of the ratios seems to be mainly influenced by the seasonal changes. This seasonal variability was assessed for the CO to CO₂ ratios for the period from February 2013 to June 2014, showing a strong anti-correlation with monthly atmospheric temperature, likely linked to seasonal changes in emissions sources (for example, domestic heating is predominant in winter and non-existent in summer). We provide evidence on the importance of residential heating in the total $\Delta\text{CO}/\Delta\text{CO}_2$ ratio. This ratio is higher than the ones for other sectors which is in contradiction to current estimates from the Airparif inventory.

The determination of these average ratios may be useful to assess the estimates provided by emission inventories. Indeed, city-scale emission inventories mainly focus on air quality, and the link with greenhouse gases, especially with CO₂, is not well made. The combination of the well-known total pollutant emissions with the ratios estimated by our experimental approach should allow a better quantification of total CO₂ emissions.

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Table 1. Observed ratios between co-emitted species derived from our method for the Multi-CO₂ campaign. Numbers in brackets correspond to 1 σ . The mole fraction ratio is reported in ppb ppm⁻¹ for $\Delta\text{CO}/\Delta\text{CO}_2$, all others to ΔCO_2 are reported in ppt ppm⁻¹. Those that do not include ΔCO_2 are reported in ppb ppb⁻¹. For readability, the diagonal (unity ratios) has been replaced by dashes (–). Ratios in bold mean that they are in a satisfactory agreement with the ones from the MEGAPOLI/CO₂-Megapolis campaign (less than 15 % of difference).

	ΔCO_2	ΔCO	$\Delta\text{Acetylene}$	$\Delta\text{Ethylene}$	$\Delta\text{Propene}$	$\Delta\text{i-pentane}$	$\Delta\text{n-pentane}$	ΔEthane	$\Delta\text{Propane}$
ΔCO_2	–	5.55 (0.24)	24.82 (2.13)	52.55 (3.87)	11.18 (2.51)	13.57 (2.34)	9.27 (0.97)	49.81 (5.10)	32.07 (2.92)
ΔCO		–	3.48 (0.28)	5.47 (0.39)	1.32 (0.08)	2.18 (0.15)	1.15 (0.11)	6.56 (0.59)	3.19 (0.30)
$\Delta\text{Acetylene}$			–	1.09 (0.06)	0.21 (0.01)	0.28 (0.02)	0.17 (0.01)	0.75 (0.10)	0.48 (0.04)
$\Delta\text{Ethylene}$				–	0.11 (0.01)	0.19 (0.01)	0.10 (0.01)	0.57 (0.04)	0.35 (0.02)
$\Delta\text{Propene}$					–	0.72 (0.04)	0.36 (0.03)	1.87 (0.20)	1.13 (0.09)
$\Delta\text{i-pentane}$						–	0.44 (0.01)	1.73 (0.11)	0.89 (0.06)
$\Delta\text{n-pentane}$							–	2.66 (0.21)	1.14 (0.08)
ΔEthane								–	0.20 (0.01)
$\Delta\text{Propane}$									–

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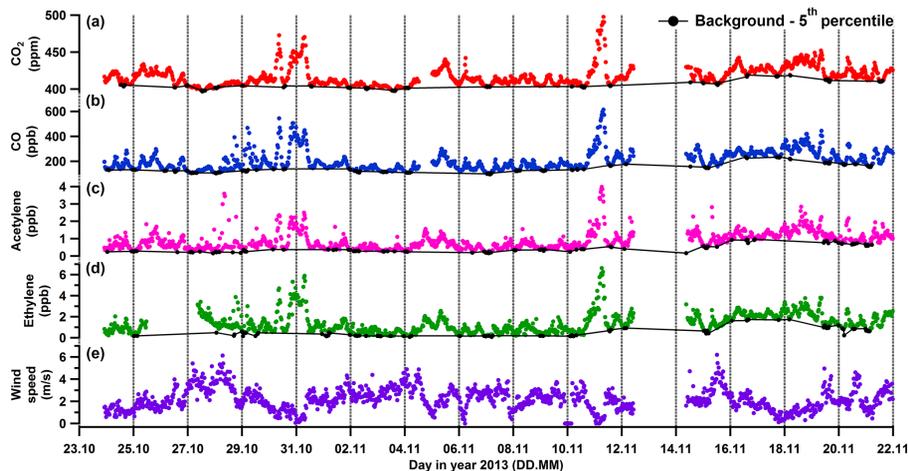


Figure 1. (a–d) Temporal variation of the mole fraction of selected compounds monitored during the Multi-CO₂ campaign (30 min time step). The black lines represent the background levels defined with the calculation of the 5th percentile (black disks). (e) Wind speed during the campaign. Time is given in UTC.

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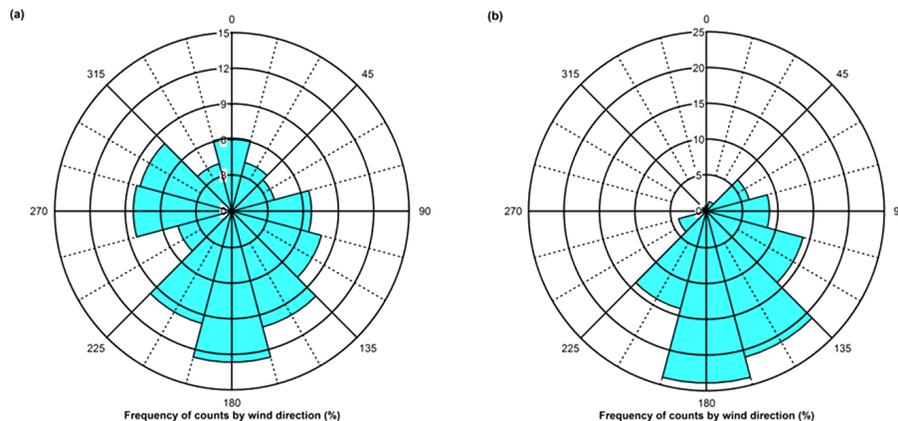


Figure 2. Wind roses for two low wind speed situations. **(a)** Wind rose for 10–11 November 2013 (significant peak in mole fractions). **(b)** Wind rose for 18 November 2013 (no significant peak in mole fractions). The percent scale is not the same for the two wind plots.

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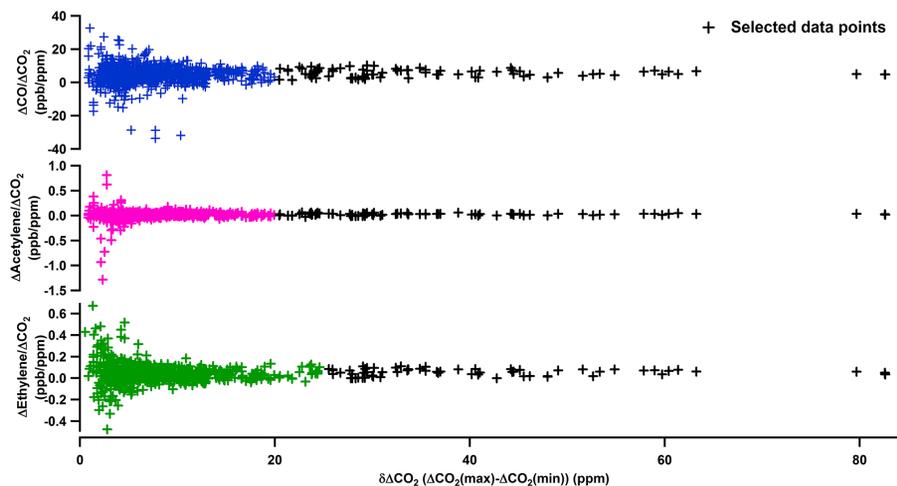


Figure 3. Selected ratios to ΔCO_2 plotted vs. the local CO_2 offset ($\delta\Delta\text{CO}_2$) from the measurements acquired during the Multi- CO_2 campaign. Black data points were selected to determine the equation of the horizontal asymptote using the criteria described in Sect. 3.3.2 (the used criteria depend on the considered species).

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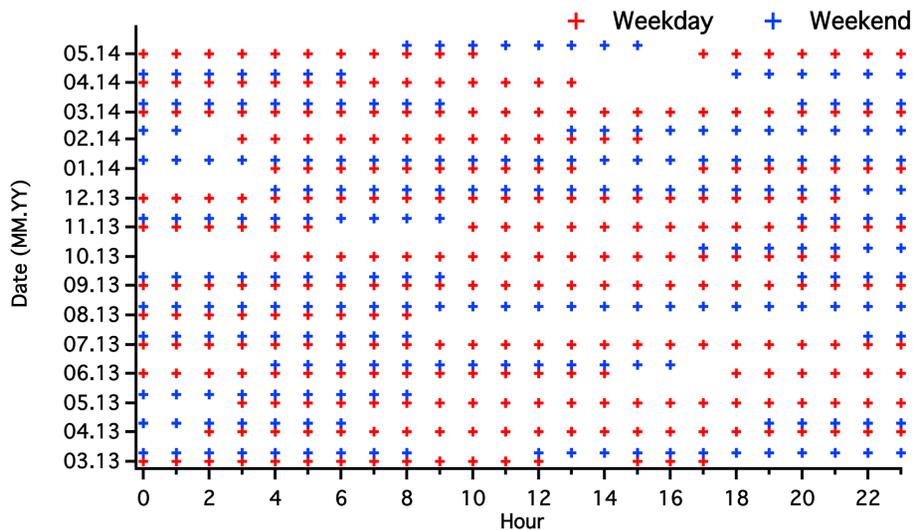


Figure 4. Days (weekdays in red crosses and weekends in blue crosses) and hour sampled per month with our method.

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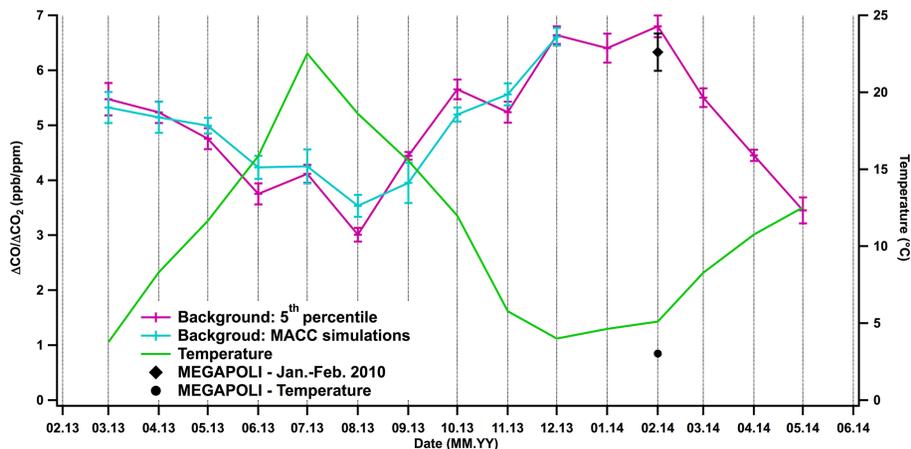


Figure 5. Monthly ΔCO to ΔCO_2 ratios in Paris. Results using background levels defined with the 5th percentile are given in violet. The ones using the MACC simulations are in blue. Error bars on the ratios correspond to 1σ . The ratio from the MEGAPOLI- CO_2 -Megaparis campaign and the corresponding average temperature are represented by a black disk. Temperature corresponding to the selected data for the ratio calculation averaged by month is represented in green as a proxy for season.