

**We thank both referees for their assessment of our paper and the useful comments. We answered to the comments and questions point by point where it is appropriate.**

**Referee #1 : Jocelyn Turnbull**

This paper describes a technique for determining emission ratios using periods of stagnant air, when mole fractions are high and therefore emission ratios can be determined more reliably than at other times. The authors show that during these periods, the choice of background is less critical than in other cases and therefore the emission ratios may be more reliable. The largest dataset is for the CO:CO<sub>2</sub> ratio, and an interesting seasonal cycle in the ratio is demonstrated.

General comments: This paper develops a good dataset and the results are quite interesting. The title and content of the paper focuses on the “new” method for estimating emission ratios, using periods of stagnant air, yet it seems a bit of a stretch to call this an entirely new method. Perhaps previous authors have not explicitly stated that they are using stagnant events in determining emission ratios, but similar methods have certainly been used.

The paper would appeal to a wider audience if the authors reduced the emphasis on the “new method”, and instead focused on the more interesting aspect – the emission ratios that they determine. The seasonal cycle in the CO:CO<sub>2</sub> ratio is discussed to some extent, but this can and should be fleshed out – how can this result be reconciled with the Airparif inventory? The VOC ratios are discussed only very very briefly and leave the reader with all sorts of questions – they could be compared to the ratios expected from emission inventories and/or from studies for other urban areas. For these reasons, I recommend major revisions to the paper. Note that the work presented appears sound, it simply doesn’t go far enough to interpret and understand the results. With revisions, the paper will be entirely appropriate for ACP.

Specific comments:

Pg 23590 lines 6-8. This sentence is phrased awkwardly. Suggest revision for clarity.

**This sentence will be rephrased.**

Section 2.2.2. Is this the same Picarro unit as used for the MEGAPOLI campaign?

**The same instrumentation (CRDS G1302, Picarro) was used for the MEGAPOLI campaign but not the same unit. The two instruments were compared in 2010 and the repeatability and the trueness of the 1 min averaged data are almost the same (listed in Lopez et al. (2013) for the MEGAPOLI campaign and in our study for the Multi-CO<sub>2</sub>-campaign).**

Lopez, M., Schmidt, M., Delmotte, M., Colomb, A., Gros, V., Janssen, C., Lehman, S. J., Mondelain, D., Perrussel, O., Ramonet, M., Xueref-Remy, I., and Bousquet, P.: CO, NO<sub>x</sub> and <sup>13</sup>CO<sub>2</sub> as tracers for fossil fuel CO<sub>2</sub>: results from a pilot study in Paris during winter 2010, Atmos. Chem. Phys., 13, 7343–7358, doi:10.5194/acp-13-7343-2013, 2013.

Please clarify what is meant by “trueness”.

**According to BIPM (2012), “measurement trueness” evaluates the “closeness of agreement between the average of an infinite number of replicate measured quantity values and a reference quantity value”. This definition will be added in the text. “Measurement trueness” is different from “measurement accuracy” which evaluates “closeness of agreement between a measured quantity value and a true quantity value of a measurand”. We will add an explanation in the text.**

BIPM : Vocabulaire international de métrologie - Concepts fondamentaux et généraux et termes associés (VIM, 3<sup>e</sup> édition), Tech. Rep. JCGM 200:2012, available at <http://www.bipm.org/fr/publications/guides/vim.html> (last access: October 2015), 2012.

Section 3.1.: first paragraph, and also in section 4.3. There is no page limit for ACP, so why not include these figures in the main paper, since they are important to the main point of the paper?

**A single example like the one of Fig. 1 appears enough to us for the main point of the paper. We prefer to keep the other figures in the supplement in order not to make the main text heavier.**

Section 3.1. Second paragraph. What VOC species were analysed? The only place they are listed is in table 1. A fleshed out discussion of the VOCs, their sources and sinks, etc should be added.

**We will list the analysed VOCs in Section 2.2.3 and describe their urban sources and sinks as well as those of CO<sub>2</sub> and CO in the introduction.**

Section 3.1. third paragraph. In the short duration stagnant air events, no buildup of mole fractions is observed. But some build-up must have occurred, just not enough to be obvious. Does the time of day that the stagnation event occurs make a difference? I suggest that the definition of a stagnant event be one where the wind is so light that the wind direction meanders. The Nov 17/18 event would then by definition be excluded.

**There is no difference in the results regarding the time of day that the meteorological events with low wind speed occurs. For example, during the Multi-CO<sub>2</sub> campaign, these events occurred during nights and days indifferently and the derived ratios are the same for all the different events.**

**We will point out the condition on the winds to be non-directional to support the assumption of “local” source. Figure 2 also illustrates this condition in the paper.**

Section 3.2. The 5th percentile baseline method does not take into account changing wind direction. For example, the lowest values could be when the wind comes from a clean air sector. When the wind comes from a sector with significant sources upwind of the city, the urban background could be much higher. How might this impact the results?

We evaluated the 5<sup>th</sup> percentile for moving windows of three days. This running window allows accounting for the dependency of synoptic situations and finally the lowest values sample indifferently the different wind sectors. To illustrate this, we present here the averages we obtained on the background concentrations we determined doing a selection on wind directions. We obtained:

- For the continental sector (0-180°), average(CO<sub>2</sub>, 5<sup>th</sup> percentile) = 410.2 ppm.
- For the oceanic sector (180-360°), average(CO<sub>2</sub>, 5<sup>th</sup> percentile) = 402.4 ppm.

This definition of the background thus accounts for different wind sectors and does not correspond to a clean air sector only.

Please add a sentence in the paragraph discussing the MACC CO<sub>2</sub> product to tell the reader that you will compare the two background methods in a later section.

**A sentence will be added to explain that the two background definitions are compared in a later section.**

Section 3.3.1. How would the results differ if the ratio was determined for each individual 30 min increment (rather than determining the slope for each 4 hour window)? The 4 hour window method seems cumbersome to calculate, whereas calculating ratios for each increment would be much more straightforward.

We focused our analyses on specific situations with low wind speed. These events appear on the different time series thanks to significant peaks in the concentrations. Finally we focused on these peaks. Our method is worthy of interest because these peaks, in which the correlations are tight, are extracted “by themselves” in the asymptote. There is no need to extract the peaks by hand. Finally, the asymptotic value for the ratios shows that these peaks can be represented by a unique ratio and this one is calculated for well-correlated data only.

As a comparison, we also determined the ratio for each individual 30 min increment. This method is easier to set up for the calculation of the ratios but the instantaneous ratios we obtained show a larger variability. For example, the instantaneous  $\Delta\text{CO}/\Delta\text{CO}_2$  ratio during the Multi-CO<sub>2</sub> campaign is  $5.91 \pm 2.61$  whereas it is  $5.55 \pm 0.24$  with our method, which confirms that our method allows a more precise determination of the ratios.

In figure 3, the asymptote appears to be  $\approx 0$  in all cases, is this a trick of the eye, or am I missing something? If the former, zero lines should be added to the graphs.

**The asymptotes are not equal to 0. This is a trick of the eye due to the scale of the y-axis. We will add the abscissa on the graphs.**

Section 4.2. second paragraph. Temperature clearly correlates to the CO:CO<sub>2</sub> ratio, but it is presumably not a direct driver, rather an indirect driver due to the possible explanations given, and - Another possible explanation for the seasonal cycle in CO:CO<sub>2</sub> ratio is that the emission ratio from traffic increases in winter. Vehicle studies suggest that the largest CO emissions occur when the vehicle starts up, and that this startup burst of emissions is larger in cold conditions (before the catalytic convertor warms up). Presumably CO emissions from other source sectors might also be higher in winter due to the lower ambient temperature.

We previously discussed the impact of the efficiency of the catalytic converter on CO emissions in Ammoura et al. (2014). CO emissions are more important when vehicles work at lower temperature than the optimal value. This optimal value is between 70 to 90°C and the time to reach this temperature is certainly longer in winter than in summer. And this excess of emissions may not be negligible in summer. To our best knowledge, no studies characterised the link between emissions and ambient temperature. Thus, we cannot rule out this possible explanation and we will add a sentence to mention this point.

Ammoura, L., Xueref-Remy, I., Gros, V., Baudic, A., Bonsang, B., Petit, J.-E., Perrussel, O., Bonnaire, N., Sciare, J., and Chevallier, F. : Atmospheric measurements of ratios between CO<sub>2</sub> and co-emitted species from traffic : a tunnel study in the Paris megacity, Atmos. Chem. Phys., 14, 12 871–12 882, doi :10.5194/acp-14-12871-2014, 2014.

In interpreting these results, the authors should consider that Miller et al (2012) showed that using total CO<sub>2</sub>, the CO:CO<sub>2</sub> ratio can be much lower than the CO:CO<sub>2ff</sub> ratio, since even in winter there can be a significant biogenic CO<sub>2</sub> source. How would the seasonality in the biogenic CO<sub>2</sub> source/sink impact the CO:CO<sub>2</sub> ratio? Could this be important to the overall seasonal cycle observed?

Our ratios are evaluated thanks to the equation of the horizontal asymptote. The data points distributed around this asymptotic value were sampled during nighttime and daytime indifferently. And for both of them, the ratio is unique (=equation of the asymptote). As night and day ratios fall into this line we may suppose that the impact of the biosphere is negligible.

The discussion of the Airparif inventory CO:CO<sub>2</sub> ratios that is in the following section would fit better here. It appears that the observed annual mean ratio is substantially higher than the Airparif inventory. Why?

According to the 2010 Airparif inventory, the annual mean ratio between CO and CO<sub>2</sub> is 3.1 ppb/ppm. Analysing our measurements, we found an observed annual mean ratio which is equal to 4.9 ppb/ppm. We thus notice 37% of difference between the two ratios.

Airparif also provided us with CO and CO<sub>2</sub> total emissions for January and August. We estimated the monthly ratios for these two months:

- CO/CO<sub>2</sub>)<sub>Jan</sub> = 3.1 ppb/ppm
- CO/CO<sub>2</sub>)<sub>Aug</sub> = 3.6 ppb/ppm

It is difficult to draw a general conclusion with only two values but it seems that there is no pronounced seasonal variability in the Airparif inventory. We will add this short comparison in the paper.

Section 4.3. First paragraph. As earlier, why not include these figures in the main paper?

Here as well we prefer not to make the main paper heavier.

As for CO:CO<sub>2</sub>, the difference in VOC:CO<sub>2</sub> ratios with temperature might be due to less efficient vehicle combustion and/or less efficient catalytic convertors in cold temperatures.

As mentioned previously, we will add a sentence in the text to allude to this possible influence.

Examining Table 1 in detail, there appear to be some inconsistencies in the ratios that should be discussed: The CO:CO<sub>2</sub> and acetylene:CO<sub>2</sub> ratios are consistent for both studies. The ethylene:CO<sub>2</sub> ratio is higher in the Multi-CO<sub>2</sub> campaign by 60%, yet ethylene:CO is very similar in both campaigns. Since CO:CO<sub>2</sub> is the same in both campaigns, this doesn't make sense! A similar situation is seen for propene and n-pentane.

We thank the referee for the careful examination of the table of ratios. Our method determined an average ratio. If we use the  $\langle \dots \rangle$  to represent an average, the ratios we determined correspond to  $\langle \frac{\Delta species1}{\Delta species2} \rangle$  and not to  $\frac{\langle \Delta species1 \rangle}{\langle \Delta species2 \rangle}$ . If we focus on the ratio between  $\Delta CO$  and  $\Delta CO_2$ , the equation of the horizontal asymptote corresponds to  $\langle \frac{\Delta CO}{\Delta CO_2} \rangle$  which is not equal, statistically speaking for two correlated variables, to  $\frac{\langle \Delta CO \rangle}{\langle \Delta CO_2 \rangle}$ . Finally, the usual simplification we can apply to derive other ratios does not work here and, for example :  $\langle \frac{\Delta CO}{\Delta CO_2} \rangle \neq \langle \frac{\Delta CO}{\Delta ethylene} \rangle \times \langle \frac{\Delta ethylene}{\Delta CO_2} \rangle$ . Therefore, it makes sense not to have this kind of mathematical links with the ratios we evaluated.

As I said in my general comments, this section is weak and would really benefit from a comparison of the observed VOC:CO<sub>2</sub> ratios with inventories and/or studies from other urban areas. There are a number of urban and regional studies that have looked in detail at the ratios of VOCs:CO that would make useful comparisons, as well as several that have looked at VOC:CO<sub>2</sub> or VOC:CO<sub>2ff</sub> ratios.

We sum up in Tables 1 and 2 hereafter some comparisons with previous studies and with the latest available Airparif inventory. Table 1 presents a comparison of ratios to CO and acetylene for the Multi-CO<sub>2</sub> and CO<sub>2</sub>-Megaparis/MEGAPOLI campaigns, and for five previous studies analyzing ratios in Paris (France), Mexico city (Mexico), New England (US) and London (United Kingdom). Estimates for the 2010 Airparif inventory are also listed in Table 1.

Table 2 exposes the comparison between the ratios we derived to  $\Delta CO_2$  and the ones referenced in previous studies to CO<sub>2ff</sub>. The comparison with the Airparif inventory is also listed.

Examining the two tables carefully, we notice important differences between all proposed ratios. Each campaign occurred in specific conditions (seasonal ratios, annual ratios), using specific background signal (or no background signal at all) and calculating ratios with different methods (using a linear regression method, instantaneous ratios). These are as many different parameters which makes it the comparison really difficult to draw general conclusions to explain all the differences we can notice.

The comparison with the latest Airparif inventory is also not completely satisfactory because we noted in Ammoura et al. (2014) that the VOC speciation matrix is out-dated

and does not account for new regulations on fuel composition for example. Furthermore, the Airparif estimates are annual ratios which are difficult to compare with monthly ratios.

Ratios	Multi-CO <sub>2</sub> (this study) – Paris – Autumn 2013	CO <sub>2</sub> - Megaparis/MEGAPOLI (this study) – Paris – Winter 2010	Borbon et al. (2013) – Paris – Summer 2009	Boynard et al. (2014) – Paris – Winter 2010	Bon et al. (2011) – Mexico City – March 2006	Warneke et al. (2007) – New England (NYC, Boston) – Summer 2004	von Schneidermesser et al. (2010) – London - 2008	Airparif 2010
$\frac{\Delta Ethane}{\Delta Acetylene}$	0.75 (0.10)	0.53 (0.03)	4.94	3.75 (0.11)	-	3.097	-	1.32
$\frac{\Delta Propane}{\Delta Acetylene}$	0.48 (0.04)	0.35 (0.02)	1.90	0.32 (0.04)	-	2.187	-	0.77
$\frac{\Delta n - pentane}{\Delta Acetylene}$	0.17 (0.01)	0.11 (0.01)	0.65	0.18 (0.01)	-	0.463	-	2.31
$\frac{\Delta i - pentane}{\Delta Acetylene}$	0.28 (0.02)	0.34 (0.02)	2.27	0.40 (0.02)	-	1.192	-	4.89
$\frac{\Delta Ethylene}{\Delta Acetylene}$	1.09 (0.06)	0.84 (0.03)	0.61	2.30 (0.05)	-	1.343	-	2.61
$\frac{\Delta Propene}{\Delta Acetylene}$	0.21 (0.01)	0.17 (0.01)	0.44	0.38 (0.02)	-	0.408	-	1.67
$\frac{\Delta CO}{\Delta Acetylene}$	287.4 (12.4)	359.7 (32.3)	242	-	-	-	-	100.02
$\frac{\Delta Ethane}{\Delta CO}$	6.56 (0.59)	3.09 (0.24)	23.4	-	21.5 (10.8)	11.616	7.1	13.17
$\frac{\Delta Propane}{\Delta CO}$	3.19 (0.30)	2.27 (0.15)	9.02	-	61.7 (15.6)	7.733	2.7	7.73
$\frac{\Delta n - pentane}{\Delta CO}$	1.15 (0.11)	0.73 (0.06)	3.08	-	2.5 (0.2)	1.548	0.54	23.13
$\frac{\Delta i - pentane}{\Delta CO}$	2.18 (0.15)	2.04 (0.11)	10.8	-	3.3 (0.4)	3.991	1.6	48.92
$\frac{\Delta Acetylene}{\Delta CO}$	3.48 (0.28)	2.78 (0.25)	4.74	-	6.5 (0.3)	3.6	-	10.00

$\frac{\Delta Ethylene}{\Delta CO}$	5.47 (0.39)	5.13 (0.33)	7.64	-	7.0 (0.4)	4.564	2.4	26.06
$\frac{\Delta Propene}{\Delta CO}$	1.32 (0.08)	0.88 (0.09)	2.07	-	3.0 (0.2)	1.363	0.72	16.74

Table 1: Comparison between ratios to CO and actetylene presented in this study and the ones provided by previous studies or by the latest Airparif inventory.

Ratios	Multi-CO2	MEGAPOLI	Turnbull et al. (2011) – Sacramento – 2009 (/CO <sub>2ff</sub> )	LaFranchi et al. (2013) – Boulder – 2009/2010 (/CO <sub>2ff</sub> )	Miller et al. (2012) – Northeast U.S. aircraft – 2004/2009 (/CO <sub>2ff</sub> )	Airparif 2010
$\frac{\Delta Ethane}{\Delta CO2}$	49.81 (5.10)	31.70 (4.41)	-	-	-	36.57
$\frac{\Delta Propane}{\Delta CO2}$	32.07 (2.92)	20.28 (2.52)	64 (18)	N/E : 3265.1 (1714.4) S : 352.7 (186.3)	138 (25)	21.43
$\frac{\Delta n - pentane}{\Delta CO2}$	9.27 (0.97)	3.41 (0.60)	18 (2)	N/E : 480.6 (151.7) S : 54.4 (31.6)	14.0 (5.4)	64.29
$\frac{\Delta i - pentane}{\Delta CO2}$	13.57 (2.34)	11.47 (1.49)	64 (8)	N/E : 485.2 (181.3) S : 65.4 (35.3)	29.5 (8.3)	135.53
$\frac{\Delta Ethylene}{\Delta CO2}$	52.55 (3.87)	33.51 (6.24)	-	-	-	72.13
$\frac{\Delta Propene}{\Delta CO2}$	11.18 (2.51)	6.26 (0.96)	-	-	-	46.33
$\frac{\Delta Acetylene}{\Delta CO2}$	24.82 (2.13)	25.21 (4.85)	52 (7)	52.1 (15.5)	34.2 (5.6)	27.69



$\frac{\Delta CO}{\Delta CO_2}$	5.55 (0.24)	6.33 (0.24)	12 (3)	7.8 (1.5)	11.2 (2)	2.20
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**Table 2 : Comparison between ratios to CO<sub>2</sub> and CO<sub>2ff</sub> presented in this study and the ones provides by previous studies and by the latest Airparif inventory**

## Anonymous Referee #2

Overview: Ammoura et al. present a manuscript showing measurements of CO<sub>2</sub>, CO and VOC's made in Paris as part of two intensive campaigns as well as longer-term observations. They analyze data in low wind conditions in what they term a new method to derive dCO/dCO<sub>2</sub> ratios with the interest of learning about emissions characteristics in the Paris region. This manuscript is well placed in ACPD. Much of the data appears sound, and the analysis pursued by the authors is worthwhile. However, I find there to be a couple key conceptual issues the authors have failed to address adequately that I will outline in detail below. Most importantly, there is a fundamental flaw in the interpretation of observed ratios as representative of emission ratios assuming dilution effects cancel between species. I outline this below, and this is a fundamental flaw that must be address. There are also some key considerations about the representativeness of observations that need more details. Finally, the authors indicate in the title and abstract that they will consider VOC's as well, but this data is largely neglected and not analyzed or discussed. Once these concerns outlined in detail below have been addressed, and VOC data have either been added into the analysis and discussion what I would encourage), I would reconsider the manuscript for publication in ACP.

**We respond point by point to these concerns in the following.**

Conceptual Issue:

Dilution-mentioned in line 38-40 "Measurements made in the ambient air are affected by dilution in the atmospheric boundary layer, but this effect cancels out when considering mole fraction ratios between the considered species." This actually is a common misconception that is not true in the cases discussed in this manuscript. This actually potentially significantly impacts the interpretation of all the analysis and requires closer examination and discussion. I can illustrate this with a simple thought experiment. Let us assume we are considering observations in Paris. Let the emissions source impacting our observation have a dCO/dCO<sub>2</sub> ratio of 5 ppb/ppm (in the normal range reported in the study here). Now the important part-let us imagine a scenario where the background CO<sub>2</sub> concentration is 380 ppm, while the free troposphere is at 390 ppm. This is just putting some simple numbers down, but this is a realistic scenario where extra-urban vegetation has drawn the boundary layer value down below the free troposphere before entering the city. Let us assume CO has 100 ppb in both the background and free troposphere. If our source emits enough CO<sub>2</sub> to raise the boundary layer by 5 ppm, then in the absence of any entrainment/dilution the observed CO<sub>2</sub> would be 385 ppm and the observed CO would be 125 ppb, and the dCO/CO<sub>2</sub> observed would match the emissions ratio of 5. Now if there is some dilution of say 25%, then the CO<sub>2</sub> measured value would be  $(.75 \times 385 + .25 \times 390) = 386.25$  and the CO measured value would be  $(.75 \times 125 + .25 \times 100) = 118.75$ . Our observed dCO/dCO<sub>2</sub> would then be  $(118.75/386.25) = 3$  - significantly different than the emissions ratio  $(25/5) = 5$ , the value we are interested in which this manuscript is attempting to measure. This is just a simple thought experiment, but clearly illustrates that dilution can change the perceived emission ratio. This is true when the background and free troposphere value are different-a situation that happens often for CO<sub>2</sub>, but also can happen frequently for CO. If

the CO<sub>2</sub> background value matched the free troposphere in the above example, we would see the dilution effects cancel. This is a critically important point that is neglected entirely in the manuscript as it is asserted in lines 38-40 that dilution effects all cancel. This is an issue the authors need to consider and include in their analysis particularly as it might have a large seasonal influence that would exactly match the seasonality reported-where the dCO/dCO<sub>2</sub> ratio drops during spring/summer. One might argue this becomes embedded in a discussion of what background value is used in the analysis, and filtering for larger delta signals lessens the impact of this concern. This would perhaps be a key place to explore this issue. Also is a place where more analysis of the VOC data could be used for further tests of this impact as for some VOC's the background value and free troposphere value will be very similar much of the time. Relatedly, using the lower 5% values may seem like a reasonable empirical choice-it almost certainly will not produce a background value that equals the free troposphere value for any of these urban sites, so could produce a bias that varies seasonally.

**We thank the reviewer for having detailed his hypothesis. However it does not fit the cases that we study. The reviewer highlights the impact of entrainment in the boundary layer. The entrainment zone is the region where the free troposphere air is mixed with the one present in the boundary layer. Even if some studies showed that this entrainment zone is thicker in urban areas, our measurements have most likely been acquired outside of this zone. For instance, Lidar measurements acquired in March 2011 in Paris (in Jussieu which is also our measurement site) showed that the boundary layer height during daytime (between 12h and 17h) was about 1135 m (Pal et al. 2012). The entrainment thickness was estimated in the same study and found to be around 400 m, meaning that more than 700 m (counted from the ground) were not influenced by the free troposphere. Our inlets were installed on the roof of a building, around 30 m above ground level and we may reasonably consider that our measurements were not influenced by the air in the free troposphere. Further, the calculation of the ratios in our method is based on the determination of the equation of a horizontal asymptote in which nighttime and daytime data points are mixed and used together for the analysis. The same ratio is thus found for nighttime and daytime data, supporting the fact that there is no influence of entrainment and free troposphere during the day in our analyses.**

Pal, S., Xueref-Remy, I., Ammoura, L., Chazette, P., Gibert, F., Royer, P., Dieudonné, E., Dupont, J. C., Haeffelin, M., Lac, C., Lopez, M., Morille, Y., and Ravetta, F. : Spatio-temporal variability of the atmospheric boundary layer depth over the Paris agglomeration : An assessment of the impact of the urban heat island intensity, *Atmospheric Environment*, 63, 261–275, doi :10.1016/j.atmosenv.2012.09.046, 2012.

Detailed Issues: Title and beyond: Calling this a 'new method' is a bit misleading, as people have studied tracer-tracer ratios extensively for decades. The tracer-tracer method has also been used in urban regions in a variety of ways; see say Wunch et al., GRL 2009 or Newman et al., ACP, 2013. There are new details in the reported approach, but it is overstating to call it a whole new method and is not needed. Generally this could be better represented in the introduction with more citations (23 references in total is a bit light and doesn't do justice to the prior tracer-tracer work done).

**The word “new” in the title does not refer to the use of the tracer-tracer method and to the determination of the ratios between the co-emitted species. It applies to the words “method for estimating emission ratios” and is therefore linked to our original approach to unambiguously compute the ratios with an asymptote.**

Title: VOC's are highlighted, but are essentially completely neglected in the manuscript.

**We present a comparison of ratios between our study and previous ones or latest estimates from the Airparif inventory as an answer to the last comment of Referee #1 (see Tables 1 and 2 in this document).**

**The comparison with the latest Airparif inventory is not completely satisfactory because we noted in Ammoura et al. (2014) that the VOC speciation matrix is out-dated and does not account for new regulations on fuel composition for example. Furthermore, the Airparif estimates are annual ratios, which are difficult to compare with monthly ratios. Thus, we are not able to compare same quantities.**

**Regarding comparisons with previous studies, we notice important differences between all proposed ratios. Each campaign occurred in specific conditions (seasonal ratios, annual ratios), using specific background signal (or no background signal at all) and calculating ratios with different methods (using a linear regression method, instantaneous ratios). These are as many different parameters, which make the comparison really difficult to draw general conclusions to explain all the differences we can notice.**

Line 15: the assessment of sensitivity to background concentration may change when dilution effects are considered.

**We have shown above that dilution effects may reasonably be neglected in our cases.**

Line 23: This conclusion would rely on the observations being representative of more than a very local site, this is not discussed or established later is of high importance.

**We will suppress the sentence.**

Line 32: This citation is not actually in the references.

**The citation was added in the references.**

Line 40-42: “.. molecules share the same origin” This is again a slight misconception that is common. The source either needs to be the same, or their needs to be sufficient atmospheric mixing between multiple sources before the observation. Subtle but important distinction.

**Our sentence refers to the interpretation of mole fraction ratios in terms of emission ratios. If atmospheric mixing merges plumes together this interpretation cannot work.**

Section 2.1: What height are the inlets? Where are the inlets related to surroundings? On a tower above the urban canopy? On a building? This is really important when considering the representativeness of the observations. Even more so than usual as looking at low wind

conditions means stagnant air might only be representative of a very small area in the direct vicinity of the observations.

**This information is already given in Section 2.1. The measurements were made on the roof of two buildings, at 23 and 15 m above ground level, respectively.**

**During the events that we considered in our study, the wind speed was lower than 1 m/s. The emission area of influence can thus be estimated to a distance of about 3.5 km around the measurement sites. Therefore, we can suppose that these conditions with low wind speed are not representative of a very small area around the station (such as the respiration impact from students going in and out the university).**

Line 175/Figure 1: The assertion that no significant peaks are visible is not true. I can clearly see a rather substantial CO<sub>2</sub> and CO feature at this low wind event. The signal is more modest than on the days of greater focus, but there is clearly very detectable enhancement there and this should be accurately represented in the text.

Line 178-179: this statement needs reassessment in light of the above comment.

**Our selection accounts also for wind directions and actually there is no specific wind direction during the events that we considered (we spoke about “turning wind”). As the first reviewer advises it, we will rephrase the definition of the events we studied, adding the criterion about the wind direction.**

Line 235: What type of linear regression is performed? Variance in both the x and y axis will be comparable so it is important to perform a regression that accounts for error in both axes (such as a Type II model regression).

**Our regression accounts for errors in both axes. We will add this clarification in the text.**

Line 285-286: This statement makes a case that perhaps the dCO<sub>2</sub> threshold choice should be defined in a way to limit the error to a certain %. If you know the ppm error, then you could define this.

**We choose to limit the error to 15% (which is the uncertainty on VOC data), keeping this way enough data points to define the asymptote and evaluate the ratio.**

Possible Biased sampling: In addition to a need to discuss the representativeness of the observations, we must also consider possible bias to the sampling. Notably, the analysis is only performed during stagnant conditions (the opposite actually of many biased samplings that only occur during sunny/well mixed conditions). What bias might this introduce?

**We have shown that the low wind speed conditions in Paris sample the hours of the day and the days of the week rather evenly, so we do not expect noticeable biases. Furthermore, our results are supposed not to be sensitive to synoptic conditions because the used background level accounts for this synoptic scale (we defined it using a moving window of three days).**

Line 324-325: It is reasonable to assess if temperature could be used as a predictor for emission ratios, but is not reasonable to consider it the driver of changing emissions ratios without establishing a physical mechanism that would explain it.

**We agree that our sentence is too affirmative. Indeed, temperature certainly has an impact on the ratios but indirectly. We will rephrase the point adding the possible influence of sources such as vehicle or heating emissions, which are more important when the temperature is low.**

Line 329-336: This is a very important paragraph, but I haven't been convinced that the analysis is actually robust to establish this paradoxical conclusion.  $dCO/dCO_2$  from emissions are expected to show the opposite seasonality reported here and there is no reason to think our notion of  $CO/CO_2$  emissions ratios from say vehicles is so grossly in error. I find it much more likely that errors in the analysis method/interpretation are better explanations for this discrepancy. Examples include: Dilution as discussed above could produce exactly the signal seen here and this is not addressed in the method. If the sampling (let's say in the Park) happens to see very strong respiration signal in spring/summer this would lower the  $CO/CO_2$ . This relates to a question of representativeness-what are the sites really representative of and what sources are in that domain? Considering only stagnant conditions are studied this may be a very small region. The authors need to establish what the size of this region may be. Representativeness needs to be addressed.

**As we mentioned previously, during the events we considered in our study, the wind speed was lower than 1 m/s and the emission area of influence can thus be estimated to a distance of about 3.5 km around the measurement sites. This area almost matches the Paris area if we consider the site of Jussieu. Our ratios may be representative of this zone and not of a very local one.**

**Regarding the differences between our measurements and the inventory estimates, we revealed the possible influence of another source. Indeed, wood burning is a major part of CO emissions from the residential sector (around 90%) but is not taken into account in Airparif  $CO_2$  emissions because it is referenced as biomass burning (and is thus not an anthropogenic component). The differences may be adjusted accounting for this source also for  $CO_2$ . However, we were not able to evaluate this point in our study but we will mention this point as outlooks of it.**