

We thank both referees for their positive assessment of our paper and the useful comments.

Anonymous Referee #1

The paper presents measurements of CO₂, CO, NO_x and VOCs from a tunnel study in the Paris region. The results are reported as ratios to CO₂, which can in principle be converted to emission factors per unit of fuel or per km driven; such measurements are important for verifying and / or updating emission inventories, like the Airparif in the present paper. Ratios of CO to CO₂ are also useful for estimating the fossil fuel burning contribution to atmospheric CO₂ variations. This paper very useful, and well suited for publication in ACP.

I find the paper very well written, concise enough and easy to follow. I recommend it for publication after minor revision. Please find my comments below.

General comments

1. The reported values for the measured species are mole fractions (or volume mixing ratios, if we consider all these gases ideal) – and not concentrations. I suggest replacing “concentration” with “mole fraction” through the paper.

We will replace “concentration” by “mole fraction”.

2. Introduction: I think some more information on VOCs would be useful: why are they important (only CO₂ tracers, or also pollution?), which are the most important species related to traffic, are they regulated by Euro standards, etc. Similarly, more background information on CO and NO_x would be good.

We will add information on VOCs, CO and NO_x in the introduction.

3. In my understanding, the night-time measurement results were averaged (for each species) and this average was then considered “background” and subtracted from individual measurements to get the Δ s. If this is correct, then the choice of background should not affect the slope of the fit (which is the ratio), but only the intercept. It is shown in the supplement that the two considered background options give the same results, but using directly the measured mole fractions without subtracting the background should give exactly the same results.

This interpretation is correct and we will make it more explicit in the revised version.

Our methodology was the following: we established the hypothesis that outside air could be brought in the tunnel, mixing different sources signals with the one of interest: traffic. To extract the traffic signal as accurately as possible, we therefore decided to remove a background signal. First, we computed the ratios using as the background signal outside ambient air concentrations (that show for CO₂ a typical diurnal cycle). However, there were only CO₂ and CO outside air data available whereas VOCs were measured together with CO₂ and CO inside the tunnel (the CO/CO₂ ratio is presented in the supplementary material).

Therefore, we decided to calculate the ratios removing an average (constant) background from nighttime measurements carried out inside the tunnel. Also note that the variability

around the mean background level (characterized by the standard deviation) is accounted for in the standard deviation given for the ratios in the tables.

The variability of the ambient measurements (diurnal cycle amplitude) was found to be around 40 ppm whereas the mixing ratios in the traffic peaks reach 900 ppm, implying an uncertainty on this method of around 4%. We found an uncertainty of the same order of magnitude by calculating the ratios with the constant background (around 5%, please see the main paper and the supplementary material). Furthermore, only minor differences were found (0.2% of difference) between the ratios calculated by both methods, which motivated the use of an average value as background.

Still, we have chosen to use the generic Δ notation in order to highlight the fact that a choice has been made about the background level definition.

4. In such a tunnel, water, gases and aerosols have higher concentrations than on open roads. How important is the chemistry in these conditions? Is it possible that some of the species measured are partly lost through chemical reactions or deposition? Or that some species are not (only) directly emitted, but formed afterwards through chemistry? In this regard, how representative would be the tunnel measurements for open air emissions?

The reviewer is right that some chemistry may occur. Concerning the VOCs, they usually react with OH radicals generated by photochemical processes that are non-significant in the tunnel conditions. In the same way the generation of secondary organic aerosols from the oxidation of VOCs involves photochemical reactions which should not be considered in this environment. Nevertheless, even the most reactive compounds measured during this campaign (xylenes, ethyl toluene) have a lifetime of several hours and therefore should not be significantly depleted within the tunnel. In addition, the good correlation of the VOCs compounds with CO and CO₂ suggests that they are of primary origin.

5. The result for fluent traffic conditions are now given as a secondary result, and only used for comparison with Popa et al., in Sect. 4.3. I see the point of the authors that these results are less well determined, and they want to report the most precise results. I think however the fluent traffic results are important and should get more attention in the paper. The emission inventories need measurement results for all traffic conditions, and imprecise results are better than no results.

We present in the following table the coefficients of determination and the ratios for fluent traffic conditions (vehicle speed > 50km.h⁻¹).

| Species | Observed ratios to ΔCO_2 | Coefficient of determination (r^2) |
|--------------------------|--|--|
| ΔCO | 5.68 ± 2.43 | 0.45 |
| ΔNO | 2.98 ± 0.68 | 0.40 |
| ΔNO_2 | 0.54 ± 0.17 | 0.26 |
| $\Delta\text{i-pentane}$ | 4.38 ± 11 | 0.0052 |
| $\Delta\text{Toluene}$ | 7.29 ± 5.06 | 0.071 |
| $\Delta\text{Acetylene}$ | 9.50 ± 5.95 | 0.077 |
| $\Delta\text{Ethylene}$ | 71.50 ± 44.2 | 0.10 |
| $\Delta\text{Propene}$ | 6.72 ± 11.70 | 0.011 |

| | | |
|--------------------------------|-------------|-------|
| Δn -pentane | 6.79 ± 11.2 | 0.012 |
| Δ Benzene | 2.04 ± 1.33 | 0.080 |
| Δm & p -xylenes | 3.50 ± 1.27 | 0.22 |
| Δo -xylene | 1.58 ± 0.83 | 0.12 |
| Δ Ethylbenzene | 0.64 ± 1.08 | 0.013 |
| Δn -propylbenzene | 0.73 ± 0.51 | 0.070 |
| Δm & p -ethyltoluene | 0.46 ± 0.21 | 0.15 |

Except for CO and NO, the correlations between Δ species and Δ CO₂ are very poor. The use of a linear regression in this case seems to be irrelevant because we cannot be sure that the sources of emissions are similar for all the species. As the aim of our study was to characterize emissions from the traffic source, we chose not to present these results.

Moreover, some of the spread of these results could be due to real variability in emissions, thus it also contains information. For CO, it is known that low speed emissions can be higher than average, even with a hot engine (e.g. Kean et al., 2003; Zhang et al., 2011). Here the comparison is made between Airparif, which I think includes average traffic conditions, and measurement results biased towards congested, low speed traffic. If all traffic conditions were considered, it could be that the authors will observe an overestimation of CO:CO₂ ratios in Airparif, related to a decrease in CO emissions over time. The CO:CO₂ ratios for fluent traffic could maybe be better estimated using the available measurements outside tunnel.

Ratios calculated for the whole campaign account for all periods of the days and the week, periods of heavy traffic (rush hours) with little or no traffic (e.g., some periods of nights or weekends), when emissions from other sources (e.g., biogenic) are brought and mixed in the tunnel. They are therefore difficult to interpret. Furthermore, the definition of the background we used might be in this case debatable.

As a complement, we present in the following table the coefficients of determination and the ratios for the whole campaign.

| Species | Observed ratios to Δ CO ₂ | Coefficient of determination (r^2) |
|---------------------------|---|--|
| Δ CO | 7.44 ± 0.15 | 0.86 |
| Δ NO | 3.99 ± 0.08 | 0.88 |
| Δ NO ₂ | 1.06 ± 0.02 | 0.83 |
| Δi -pentane | 27.84 ± 1.01 | 0.67 |
| Δ Toluene | 21.99 ± 0.70 | 0.71 |
| Δ Acetylene | 14.65 ± 0.56 | 0.64 |
| Δ Ethylene | - | - |
| Δ Propene | 10.37 ± 0.62 | 0.42 |
| Δn -pentane | 9.49 ± 1.59 | 0.085 |
| Δ Benzene | 7.54 ± 0.19 | 0.80 |
| Δm & p -xylenes | 4.84 ± 0.14 | 0.74 |
| Δo -xylene | 3.46 ± 0.10 | 0.74 |

| | | |
|---------------------------|-----------------|------|
| Δ Ethylbenzene | 2.68 ± 0.08 | 0.71 |
| Δ n-propylbenzene | 2.60 ± 0.10 | 0.71 |
| Δ m&p-ethyltoluene | 1.35 ± 0.05 | 0.64 |

According to this table, the correlations between the co-emitted species and CO₂, considering all the data of the campaign, are good (except for n-pentane). However, we notice that these ratios are lower than the ones evaluated in the traffic peaks. The differences with the ratios provided by the Airparif inventory are even more important.

Abstract, line 12: I suggest to replace “and rush hour periods” with “and on rush hour periods”

Abstract, line 13: I think “To those ...” should be “From those ...”

Abstract, line 16: “ Δ species” – later in the paper is “ Δ Species”, with capital S – check consistency

page 20199, line 25: I think “characterized” should be “characterize”

page 20199, line 26: the word “well” should be moved at the end of the phrase, or after “represent”

page 20200, line 1: “Megaparis” is spelled sometimes “MegaParis” – please check consistency

page 20200, line 5: “and of its carbon isotopes” – I would remove “of”

page 20200, lines 8 – 9: “to originate for 30% from traffic and for 70% from gas heating” – I think the two “for” should be removed

page 20203, line 8: “analysed continuous CO₂, CO and H₂O measurements” – I suggest to replace with “performed continuous CO₂, CO and H₂O measurements” or “analysed/measured CO₂, CO and H₂O continuously”

page 20203, line 11: “Gas Chromatograph” does not need capitals

All these corrections will be done.

page 20203, line 11: please check the calibration scales: by my knowledge NOAA-X2007 scale is only for CO₂, and the most recent calibration scale for CO is X2004.

We checked the calibration scale and agree with this comment. We will correct the text.

page 20203, line 22: it would be good to mention here which NMHC species were measured

The list of NMHC species we measured will be added.

page 20204, lines 3 – 4: “The total uncertainty on the data was estimated better than 15%” – please consider reformulating, e.g. “The total uncertainty of the data was better than 15%”

This will be corrected.

page 20208, lines 20 – 25: Are the species emitted from fuel evaporation not correlated to the number of vehicles, the same as CO₂? If yes, shouldn't there be a better correlation between these species and CO₂? Also, did these species exhibit higher than background

mole fractions in the tunnel? – if yes, and if there are not many such measurements published, it may be useful to report them, at least as a time series plot in the supplement.

We studied the correlations between the species and the number of vehicles. We found very poor coefficient of determination ($r^2 < 0.1$), especially as regards species emitted from fuel evaporation ($r^2 < 0.01$). We finally decided not to present the results about these species.

page 20209, lines 13 – 15: Did Gros et al., 2014 use the same data? If yes, then it's not surprising that the results are similar.

The same data are used for this study. We will rephrase the sentence.

page 20216, line 25: "Volatile" typo

This will be corrected.

page 20219, lines 29 – 31: Is Roustan et al. still in press?

The updated reference is : Roustan, Y., Pausader, M., and Seigneur, C.: Estimating the effect of on-road vehicle 30 emission controls on future air quality in Paris, France, *Atmos. Environ.*, 45, 6828-6836, doi:10.1016/j.atmosenv.2010.10.010, 2011.

page 20222, Table 1 caption: there is no ΔNO_x in table, but ΔNO and ΔNO_2

This will be corrected.

page 20223, Table 2: I would also include here on a separate line the results of this study for fluent traffic. See also the general comments.

Please see the answer to the general comments.

Reviewer #2 : Jocelyn Turnbull

This paper describes a tunnel study measuring a suite of anthropogenic trace gases. They use the ratio of each gas to CO₂ to evaluate the emission rates of these gases and compare with bottom-up inventory data. The authors compare their results with other studies in other urban areas, and provide some explanations for the similarities and differences. Of particular interest is the detailed analysis of the CO/CO₂ ratios, which highlights the variability in this ratio, and as the authors point out, makes using CO measurements as a proxy for fossil fuel CO₂ a real challenge

This paper was really a pleasure to read. The topic is clear, the experiment well designed, the analysis is well explained, and the discussion nicely covered all my initial questions. The subject material is entirely appropriate for ACP. I have a few very minor comments on points of clarity, but the paper could be published as is without problems.

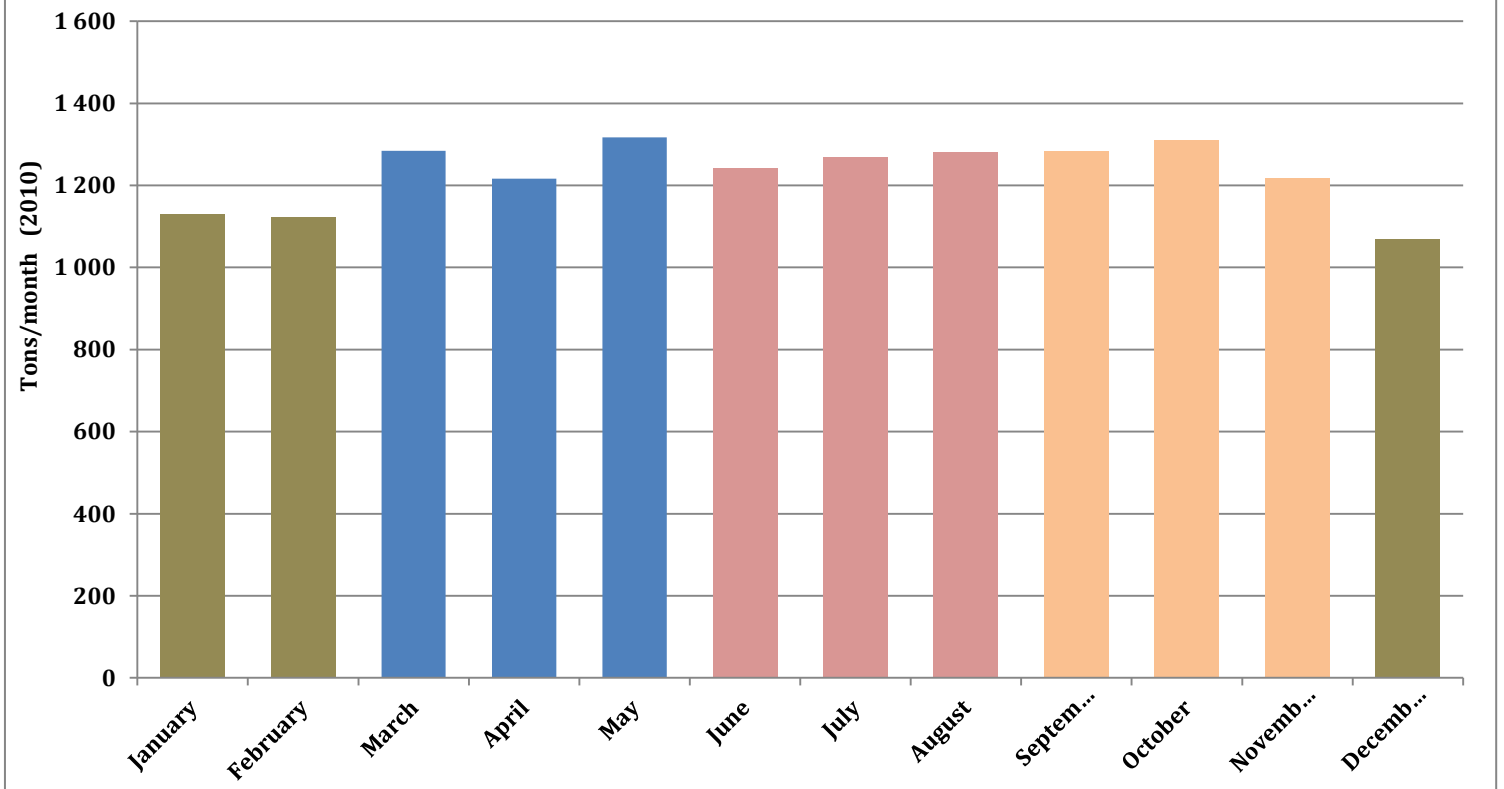
Pg 20199 Line 20. What is meant by “tertiary sector”?

According to the Airparif report (Airparif, 2013), the tertiary sector groups together several types of activities such as hotels, restaurants, bars, offices of companies, health and social care institutions, community and sports centers, schools, ... The emissions taken into account for this sector are energy consumption linked to these activities. In order to be clearer, we will replace “tertiary sector” by “service sector”.

Pg 20211. Lines 1-4. Are the emission ratios also temperature dependent (more VOC and CO emissions when air temperature and/or engine temperature is lower)? If so, do September and October approximate the annual average temperature for Paris, or might we expect that the observations could be biased relative to the annual average Airparif inventory?

Emissions ratio may be slightly influenced by temperature but this effect could not be investigated in this study. Nevertheless, we do not think this effect could bias our comparison with the emission inventory. According to the Airparif inventory, there is indeed a small seasonal variation in the VOCs emitted by traffic. Nevertheless, September and October contributions to the whole year are close to the yearly average and therefore can be considered as representative. This point is shown on the following figure, which represents average emissions of VOCs per month considering the whole traffic sector (exhaust and evaporation).

WHOLE SNAP 07 (ROAD-TRAFFIC)



(Airparif 2010 inventory. We thank Alexia Baudic for this figure).

Pg 20211 line 28. Table 3, not table 6.

This will be corrected.

Pg 20213 line 25-27. This last sentence of the paragraph is unclear.

We will rephrase the sentence.

Pg 20214 lines 7-16. The most likely explanation is the first one given – at higher speeds, the CO:CO₂ ratio is lower. The further explanation regarding tunnel ventilation seems less likely, but the phrasing of the paragraph seems to overly emphasize this second possibility. I would rephrase the second explanation to something like “we cannot rule out the possibility that. . .”

We will follow this suggestion.

Figure 1. It would be nice to see a larger version of this key figure, as it is difficult to examine hour-by-hour at this resolution. Although not the main intent of the paper, the observed diurnal cycles in the mixing ratios will likely be of interest to many readers.

We will put an improved version of the figure in the revised paper and add comments on the mean diurnal cycles in the mixing ratios and their variability.