RC2: 'Comment on essd-2025-602', Anonymous Referee #2, 23 Dec 2025

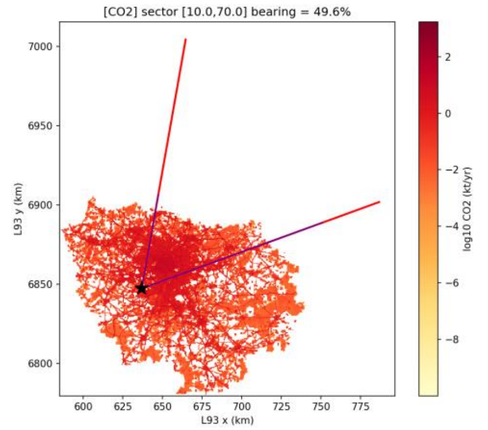
This manuscript presents a comprehensive analysis of a ten-year dataset (2012–2022) of atmospheric composition measurements at the Saclay/SIRTA peri-urban observatory near Paris. The study is timely, given the increasing importance of quantifying urban greenhouse gas and pollutant emissions, and the dataset itself is a significant contribution to the research community. The paper is generally well-structured, methods are clearly described, and the use of wind-sector separation to derive an "urban offset" is a sound and innovative approach for attributing concentration enhancements to Paris emissions. The co-location of greenhouse gas and pollutant measurements within the ICOS and ACTRIS infrastructures adds considerable strength to the analysis. However, several aspects require clarification, expansion, or correction before the manuscript can be considered for publication.

1. The choice of wind sectors for defining "urban" (10–70°) and "rural" (190–280°) conditions is central to the analysis but is not sufficiently justified.

- **Why were these specific angular ranges selected? A map showing the location of major emission sources (Paris center, other towns, major roads) relative to these sectors would greatly help readers assess whether the sectors truly isolate "Paris influence" vs. "background".**
- **The wind-speed filter of >4 m s^{-1} for the rural sector is applied to "avoid the impact of nearby cities", but no such filter is applied to the urban sector. Could low wind speeds in the urban sector lead to contamination from very local sources not representative of Paris outflow? Please justify this asymmetric treatment or consider applying a consistent wind-speed threshold.**

Regarding the first point, as suggested, we have added a map showing anthropogenic CO2 emissions in the Ile de France region, as well as the angular sector that was selected for our study. Le secteur retenu contient 50% des émissions anthropiques de l'inventaire Airparif. The sector was chosen to include the areas with the highest density of anthropogenic emissions. Extending the southern boundary of the sector would allow for the inclusion of more emission-generating areas, but the sector would also include forested or agricultural areas, shown in white on the map, which would interfere with the information sought regarding anthropogenic emissions. We have added the figure in the supplementary material (Figure A3), as well as this comment in the main text:

"*For the urban sector, the selected wind directions are defined to be between 10◦ and 70◦, corresponding to the north-easterly direction (Figure 1, Figure A3). The boundaries of the urban area were chosen to include the zone with high densities of anthropogenic emissions, while minimizing as much as possible the inclusion of forested and agricultural areas that would interfere with the anthropogenic signal of interest.*"



A wind speed of 4 m/s was selected for rural areas in order to minimize the influence of the cities west of the station. At this speed, the air mass traveled 14.4 km in one hour, which is the data integration time we use. Given that the Saclay station is only 20 km from the city center of Paris, we concluded that applying the same wind speed filter would effectively eliminate the influence of anthropogenic emissions over most of the distance between Saclay and Paris. One alternative, which has not been developed, would be to filter the minute-by-minute measurements before calculating the hourly averages

2. The authors mention that data gaps >30 days are not interpolated in the smoothing procedure, but it is unclear how this affects the trend analysis, especially for compounds with shorter records or more frequent

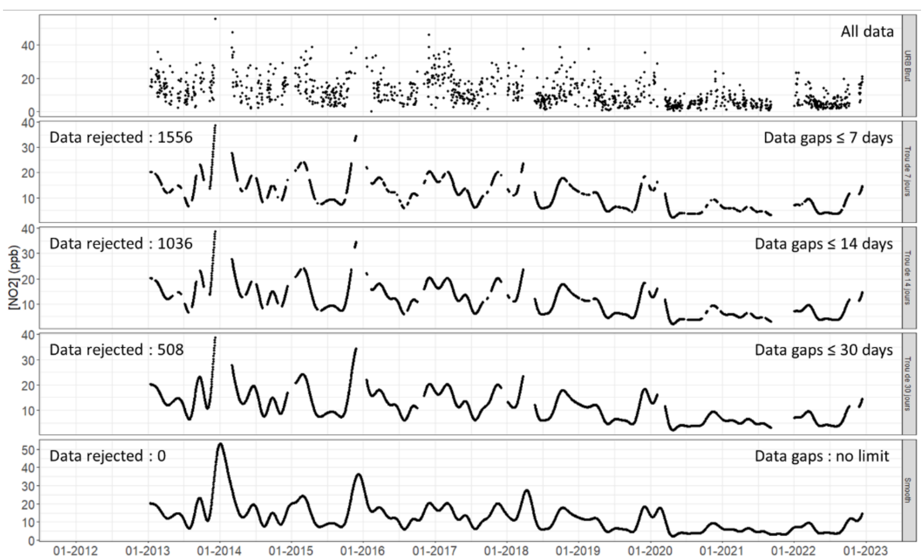
gaps. A supplementary figure showing data coverage (hourly/monthly) for each species in each sector would be helpful. The instrument inter-comparisons (GC vs. Picarro) in Appendix A are reassuring, but the text notes that values for some compounds are as low as 0.84. While slopes are near 1, some scatter is evident. A brief discussion on how this measurement uncertainty propagates into the urban offset calculation—especially for species with small offsets—would strengthen the methods section.

We did indeed believe that it would be risky to interpolate the measurement series over periods that are too long. We have been testing interpolation over periods from 7 to 30 days, or without any limit. The differences in the fitted curves were quite small, as shown by the two examples below for NO2 and CO2, both from urban sectors. That said, interpolating all the data gaps seemed to exaggerate certain peaks, such as the NO_2 peak in January 2014, with values exceeding 50 ppb during a fairly long data gap. The 30-day period therefore seems like a good compromise between interpolation and rejected events. The 2 figures below have been added in the supplementary material (Figure A4).

As suggested we have also added one figure in the supplementary (Figure A5) to show the data coverage for all species, in rural and urban sectors.

The relatively low correlation of 0.84 corresponds to methane. The trend in methane concentrations is consistent with that observed at stations in the global network, and since we do not observe a significant trend in the urban CH_4 offset, this long-term signal is not discussed in the paper.

NO_2 URB



CO_2 URB

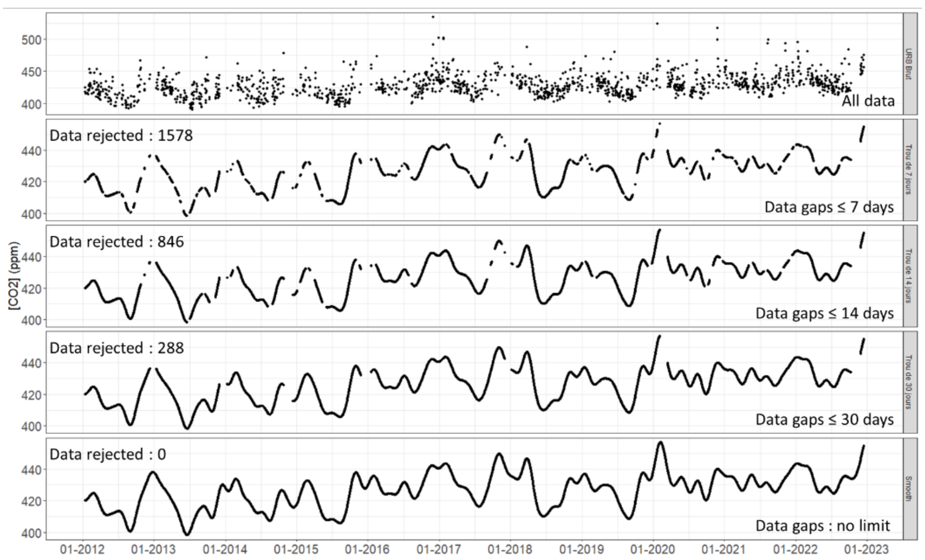


Figure A4. Comparison of the fitted curves obtained for NO_2 and CO_2 in urban sectors, after interpolating all data gaps (lowest panel), or only data gaps less or equal respectively to 30, 14, or 7 days. The total number of days, when interpolating all the data gaps, is 3991 days. The number of days excluded for each case is shown on the left.

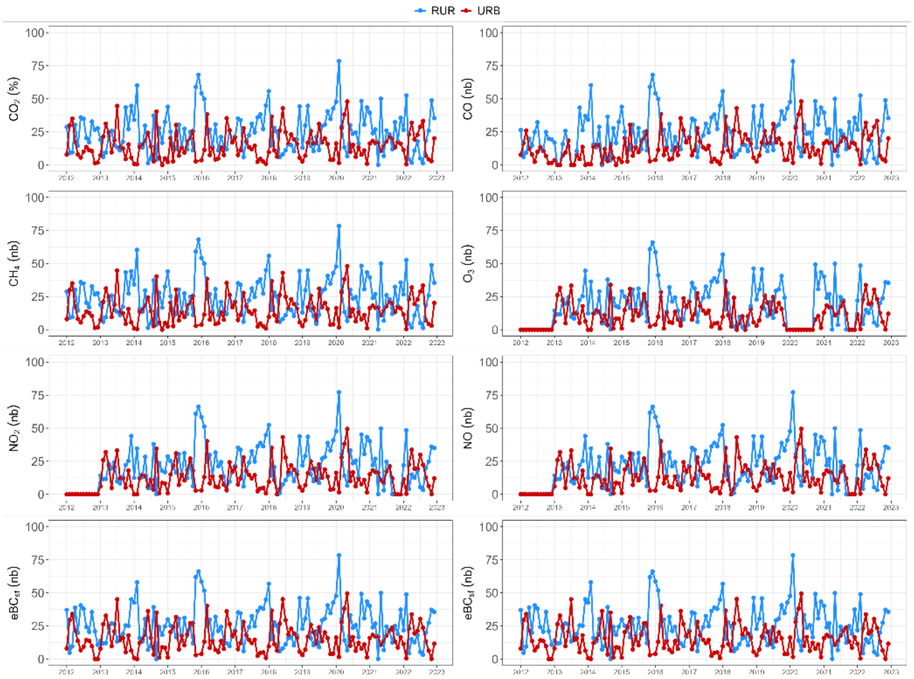


Figure A5. Month-by-month percentage of data for different species in rural (blue) and urban (red) sectors.

3. The 15% reduction in the CO_2 urban offset between 2012–2017 and 2019–2022 is highlighted as consistent with Airparif inventories. However, the strong influence of biospheric fluxes on CO_2 is acknowledged elsewhere in the text. How were inter-annual variations in biospheric uptake/respiration (e.g., due to climate anomalies) accounted for when comparing these two periods? Could such variability partly explain the observed "break" between periods? A more detailed discussion on the challenges of attributing CO_2 changes solely to anthropogenic emission trends is needed.

It is very clear that biospheric fluxes play a significant role in the observed concentrations, as well as in their seasonal and diurnal cycles. With regard to decadal trends, it is indeed challenging to determine the extent to which ecosystems may be contributing, and we have included a note regarding this difficulty in the text. One piece of evidence comes from the ICOS forest station at Fontainebleau-Barbeau (https://meta.icos-cp.eu/objects/FRqdGz1Hn6ANHW8o2e_hsv4c). Eddy-covariance flux measurements taken at this site since 2006 show no significant trend. A significant variation is observed in 2023, but only due to logging within the tower's footprint. We have added in the paper the following text in the conclusion section:

"Atmospheric CO2 measurements result from the combined contributions of anthropogenic emissions and biospheric fluxes (uptake and respiration). Therefore, a biospheric contribution to this observed signal cannot be completely ruled out. However, CO2 flux measurements by eddy-covariance taken at the Barbeau ICOS site south of Paris, in the Fontainebleau Forest, do not indicate a significant trend over the 2006–2025 period. A significant variation is observed in 2023, but only due to logging within the tower's footprint (https://meta.icos-cp.eu/objects/FRqdGz1Hn6ANHW8o2e_hsv4c)."

4. The comparison between atmospheric ratios ($\Delta CO/\Delta CO_2$, $\Delta NO_x/\Delta CO_2$) and inventory ratios is interesting, but the discussion is somewhat cursory. The manuscript states that atmospheric ratios are "lower than the ratio deduced from the inventory" and suggests biospheric fluxes as a possible explanation for CO_2. For NO_x, the reactivity of NO_x during transport is mentioned, but could there also be a systematic underestimation in the inventory's spatial allocation or temporal profiles? A more quantitative exploration of these discrepancies (e.g., using a simple dispersion model) would elevate the discussion.

As you mentioned, in our discussion, we consider the impact of biospheric CO2 fluxes, which are not accounted for in the inventories and therefore automatically contribute to lower ratios in the inventories. Then, the reactivity of NOx is also discussed as one of the possible causes of the differences in the ratios. We agree that inventory errors may also play a role, and we have added a sentence to that effect in the text. The challenge is that inventories do not have associated uncertainties, and using a transport model to test the influence of various errors on the intensity and spatiotemporal distribution of emissions seems to us to go beyond the scope of this study.

"*Figure 7 shows that atmospheric ratios of the urban offset are lower than the ratio deduced from the inventory, both for CO/CO2 and NOx/CO2 ratios. We cannot rule out the possibility that this difference may be partly attributable to inventory errors for one or the other of the compounds. In the absence of information on inventory uncertainties, it is difficult to explore this hypothesis. On the other hand, the fact that CO2 biosphere fluxes are not accounted for in the inventories, but have a significant impact on the atmospheric CO2 concentrations, inevitably plays a role in the lower CO/CO2 and NOx/CO2 ratios of the inventory.*".

5. The methodology for eBC source apportionment (Sandradewi et al., 2008) is described, but the chosen values of αsf and αlf (1.85 and 0.9) are critical. Were these values validated for the Paris region? A brief justification or reference to local studies would be helpful. Additionally, the trends in $eBC_s f$ (increase since 2020) are noted, but the discussion would benefit from linking this more explicitly to reported changes in wood-burning practices (e.g., energy cost increases).

We have supplemented the description of the BC measures with the following text and associated references:

"*The reliability of BC source apportionment results mainly depends on the alpha values used for liquid-fuel and solid-fuel fractions (Favez et al., 2010). Navarro-Barboza et al. (2024) estimated an overall uncertainty not less than 20%. Given the extensive statistical work carried out at SIRTA regarding BC source apportionment (eg Savadkoohi et al., 2025) leading to robust and consistent couple of alpha values, a 20% uncertainty can be applied for BClf and BCsf.*"

It seems difficult to us to go any further in analyzing recent trends in the wood industry in the Île-de-France region. There are reports indicating a significant increase in wood use, but this has been accompanied by the modernization of facilities, leading to a reduction in particulate emissions. At this point, we have not been able to find any publications on trends in the wood-burning practices in the Île-de-France region.

"*With regard to eBCsf from solid fuel combustion, mainly associated with biomass burning, a slight increase in concentrations has been noted since 2020, which could be driven by an increase in wood-burning emissions due to climate policies and rising energy costs (Savadkoohi et al., 2023).*"

Favez, O., El Haddad, I., Piot, C., Boréave, A., Abidi, E., Marchand, N., Jaffrezo, J.-L., Besombes, J.-L., Personnaz, M.-B., Sciare, J., Wortham, H., George, C., and D'Anna, B.: Inter-comparison of source apportionment models for the estimation of wood burning aerosols during wintertime in an Alpine city (Grenoble, France), Atmos. Chem. Phys., 10, 5295–5314, https://doi.org/10.5194/acp-10-5295-2010, 2010.

Navarro-Barboza, H., Pandolfi, M., Guevara, M., Enciso, S., Tena, C., Via, M., Yus-Díez, J., Reche, C., Pérez, N., Alastuey, A., Querol, X., and Jorba, O.: Uncertainties in source allocation of carbonaceous aerosols in a Mediterranean region, Environment International, 183, 108252, https://doi.org/10.1016/j.envint.2023.108252, 2024.

Savadkoohi, M., Gherras, M., Favez, O., Petit, J.-E., Rovira, J., Chen, G. I., Via, M., Platt, S., Aurela, M., Chazeau, B., De Brito, J. F., Riffault, V., Eleftheriadis, K., Flentje, H., Gysel-Beer, M., Hueglin, C., Rigler, M., Gregorič, A., Ivančič, M., Keernik, H., Maasikmets, M., Liakakou, E., Stavroulas, I., Luoma, K., Marchand, N., Mihalopoulos, N., Petäjä, T., Prevot, A. S. H., Daellenbach, K. R., Vodička, P., Timonen, H., Tobler, A., Vasilescu, J., Dandocsi, A., Mbengue, S., Vratolis, S., Zografou, O., Chauvigné, A., Hopke, P. K., Querol, X., Alastuey, A., and Pandolfi, M.: Addressing the advantages and limitations of using Aethalometer data to determine the optimal absorption Ångström exponents (AAEs) values for eBC source apportionment, Atmospheric Environment, 349, 121121, https://doi.org/10.1016/j.atmosenv.2025.121121, 2025

6. Abstract: "$eBC_l f$" is used without definition; please spell out "equivalent black carbon from liquid fuel" on first use.

All variables names, including eBC_{lf} have been written in full in the abstract.

7. Introduction: The sentence "Atmospheric studies have endeavoured to estimate urban CO_2 emissions. The study by Levin et al. (2011) have been analyzed…" contains a grammatical error ("have been" → "analyzed"). Please rephrase.

The sentence has been corrected. Thanks for noticing the error.

8. In Figure 5 (diurnal cycles), indicating the time axis in local time (CET/CEST) alongside UTC would improve readability for a regional audience.

Thank you for pointing out this inaccuracy. The correction to Figure 5 has been made.

9. The data DOI is provided, which is excellent. It would be helpful to also mention the expected availability date (upon publication?) and whether boundary-layer height data are available through another repository.

The measures presented in this paper are already available at this link.

10. Line 185: It is interesting to see O3 levels were higher in rural sector than urban sector. It could be transported precursors from nearby cities.

We agree with the reviewer that precursors can be transported from nearby cities, including from Paris. The following paragraph has been added to the revised version

"We note that for O_3, a secondary pollutant, the rural sector shows higher concentrations because of the complex mechanics of its formation. *Indeed, as the ozone formation depends non-linearly on the interaction between NOx and VOCs and because urban areas are typically NOx-rich but VOC-limited while rural or peri-urban areas are often VOC-rich and NOx-poor, the highest ozone concentrations can frequently occur in rural regions downwind of urban emissions. In the Paris region, these are on average oriented along a northeast - southwest axis, transporting precursors and creating conditions where an optimal VOC/NOx ratio enhances photochemical ozone production (F. Gaie Levrel, Airparif, person. comm).*"

11. Typos and formatting:

- **Page 2, line 55: "process tracers," → "process tracers" (remove comma).**
- **Page 7, Table 2: "Tertiaire" should be "Tertiary" for consistency. The table caption mentions "BC" but the table header says "BC"; please verify.**
- **Page 12, line 235: "As mentionned above" → "As mentioned above".**

Thank you for pointing out these errors; we have corrected them.