RC1: 'Comment on egusphere-2024-2826', Anonymous Referee #2, 07 Oct 2024
Review of "The monitoring network of greenhouse gas (CO2, CH4) in the Paris' region" by Doc et al., for Atmospheric Chemistry and Physics (ACP).

This manuscript provides a comprehensive overview of the greenhouse gas (GHG) monitoring network in the Paris region, including site descriptions, calibration strategies, data quality assurance, and observed trends from 2015–2022. The long-term, high-frequency dataset is a valuable contribution to urban GHG monitoring and modeling efforts. The manuscript is detailed, but the analysis and presentation could benefit from further clarification and refinement. I recommend publication after attention to the items below.

<u>Overall comments:</u>

1. Several papers describing aspects of this network have been published previously. A more comprehensive summary of what is already in the literature and what is new here would be helpful. Also, references to other networks should be updated to include the most recent work on inversions and network setup.

In response to your point, we have added references to recent publications (2025) on urban networks in South Korea and North America. We have also added a paragraph describing the current atmospheric network in Paris and the differences from previous publications. We have also added the first analysis of the ten-year trend in the urban dome of CO_2, CH_4, and CO in Paris.

"*Urban atmospheric GHG measurement networks have been deployed in few cities (Mitchell et al. 2022, Verhulst et al. 2017, Gonzalez Del Castillo et al. 2022, Dietrich et al. 2021, Pugliese et al. 2018, Shusterman et al. 2016, Lian et al. 2024, Lauvaux et al. 2020*, *Kim et al.* 2025, *Sim et al.* 2025*) for estimating emissions based on a methodology referred to as atmospheric inversions. Among the CO2 measurement networks in urban areas, we can distinguish between measurements taken with highly accurate instruments, similar to background networks, and networks based on so-called mid-cost instruments, which aim to achieve measurements with an accuracy of around 1-2 ppm, whereas accurate instruments achieve an accuracy of 0.1 ppm. Dense networks of mid-cost instruments have proven effective in assessing high variability of atmospheric CO2 in city centers (Kim et al., 2025). Here we present the results of a measurement network covering the Ile-de-France region, which is unique in terms of its duration and measurement density using precise analyzers. In total, eight years of continuous, high-precision measurements of CO_2, CH_4, and CO concentrations were carried out by a network of seven ground stations located around and within the city of Paris.*"

"*We also present the first analysis of the ten-year trend in CO_2, CH_4, and CO offsets above the city of Paris.*"

"*A first greenhouse gases monitoring network was initiated in 2010 in the Ile de France region with 5 stations (Xueref-Remy et al., 2018). Of this network, only one station (Jussieu-QUALAIR) has been maintained to date, but in 2010 only PBL measurements were available. The Eiffel Tower (EIF) and Montgé-en-Goelle (MON) stations had to be shut down. The other two stations of the 2010 network had to be relocated to new infrastructures. Gonesse (GNS) station has been moved from a fire station in the city centre (4m agl) to the top of a water tower 2.5 kilometers away (36m agl) on the edge of the city. Finally, the initial GIF station located on the roof of the LSCE building (7 meters agl) was shut down in favor of the ICOS class 1 station in Saclay 1.5 kilometer away), with air sampling at 15, 60 and 100m agl. The measurement network of greenhouse gases in Île-de-France region is currently composed of seven stations: the two first stations (OVS and COU) have been set up in 2014 and the last one (GNS) has been set up in 2017. CDS and JUS stations are considered as "urban sites" due to their location inside the Paris city. The five remaining sites are located around Paris and are called "peri-urban sites" (Fig. 1). Among these, Saclay (SAC) station is part of the European ICOS network (Heiskanen et al. 2022).*"

J.Kim, W. M. Berelson, N. E.Rollins, N. G. Asimow, C. Newman, R. C. Cohen, J. B. Miller, B. C. McDonald, J. Peischl, and S. J. Lehman, Environmental Science & Technology, 59 (7), (2025), pp. 3508-351

Sim, S. and Jeong, S.: Constraining urban CO2 emissions in Seoul using combined ground and satellite observations with Bayesian inverse modelling, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2025-3367, 2025.

2. The authors should also consider whether their assumption that CH4 and CO do not have the same source is justified by the data. I agree that inventories widely assume that the two have different sources in cities. However, these results suggest methane emissions are proportional to CO and therefore leaks are proportional to consumption.

We have revised the paragraph concerning the correlations between CO and CH_4 to take into account the two reviews of the manuscript.

"*In cities, methane is mainly emitted by waste management (landfills and wastewater) and leaks in urban gas distribution network (Defratyka et al., 2021). In these two cases, there is no co-emission of CO.* ~~*Bio-fuels may be responsible for co-emission of CO and CH_4, but this remains a minority.*~~

…

According to Airparif's inventory, the main sources of CH_4 in the Ile-de-France region are the waste sector (60%) and the residential sector (20%). For CO, the residential sector dominates (67%), followed by road traffic (20%). Some emissions are therefore common to both gases, but in the case of CH_4, waste disposal sites located on the outskirts of cities represent a major contribution. The difference in emissions is reflected in the diurnal cycles (Figure 8), with a double maximum linked to rush hours that is more pronounced for CO than for CH_4. Even though emissions result from different activities, they can be co-located at the station footprint scale, which explains the correlations between the two gases, which are affected by the same atmospheric dynamic processes.

~~*As methane sources are different from CO sources, it was not expected to find such a strong correlation between these two species. We assume that the sources of methane and carbon monoxide are colocated in the footprint of the stations, but that they are not identical. The correlation between these two species is due to the colocation of these sources and to the dynamics of the atmosphere that affects all the air mass. In order to investigate this possibility, we will study the emission maps for these two species and determine the extent to which the sources of CO and CH_4 coincide (or not) in a future work."*~~

3. Identification of Plumes:

Both of the filtering methods need to be explained more clearly. While the authors refer to a "statistical filter" derived from high-frequency variability (citing El Yazidi et al., 2018 and Cristofanelli et al., 2023), the manuscript does not provide a self-contained summary of how this filter works. Since this filtering step is critical to all subsequent analyses including diurnal cycles, covariability, and gradients, it should be briefly described in the main text. Specific suggestions:

The paper should provide a brief description of the statistical method (e.g., moving median, standard deviation thresholding, percentile ranges). What thresholds are applied? 2) Clarify which types of anomalies the filter is designed to catch (e.g., define short spikes clearly). How does the filter determine the baseline concentration levels? 3) Is the same filter applied uniformly to all species and stations, or are there species-specific thresholds or site-specific tuning? 4) Was the output of the statistical filter visually checked, or compared to known events (e.g., maintenance logs or known pollution spikes)?

The request for a brief description of the statistical method was also a request from the first reviewer. We have added a description of the method, with the values of the parameters used in this study. The same parameters have been used for all stations, without any site-specific tuning. The effectiveness of the statistical method was evaluated as far as possible at different sites by Cristofanelli et al. 2023. In the case of Saclay, the method is effective in identifying spikes linked to emissions from the heat gas plant located 800 m from the measurement site.

"*The spikes targeted by this approach are variations lasting a few minutes caused by strong emission sources located within few kilometers from the monitoring station. This method consists of calculating the standard deviation (σ) of data falling between the first and the third quartile of the entire dataset. Spikes are defined by data values higher than a threshold defined as the concentration of non-spike concentration plus a threshold* *value value: α × σ + (n)$^{1/2}$ × σ, where α is a parameter to control the selection threshold, and n is the number of points between the non-spike value and the evaluated minute concentration. Based on sensitivity tests, α is set to 1 for CO_2 and CH_4 and to 3 for CO (El-Yazidi et al., 2018). The same parameters were used for all stations."*

The wind sector filter approach to identifying and removing local contamination at GNS and OVS is interesting but currently underdeveloped. The manuscript should provide enough information for someone to repeat the analysis and arrive at the same result. The description states that filter thresholds for wind direction, wind speed, and standard deviation were "determined empirically," but no clear procedure or rationale is provided. Were these thresholds optimized? Consider describing the steps taken to identify thresholds and validating them (e.g., correlation drops, concentration anomalies, footprint analysis). The authors mention that 5–13% of hourly data are flagged as contaminated using either of data filters but didn't show any quantitative analysis results, which leads to the next question: how does the wind sector filter compare with the high-frequency spike filter used network-wide? Are there overlapping selections? A brief comparison of data removed by each filter and whether one dominates would improve clarity.

The two filters apply to different data aggregates (minute for the statistical filter, hourly for the wind sector filter) and are therefore not easily comparable point by point. However, we have included the following text comparing in detail the impact of these two filters at the GNS and OVS stations. It appears that in the case of OVS, both methods filter mostly the same data, whereas at GNS, the wind filter does not effectively eliminate spikes, probably due to several sources of local contamination. Here is the additional information added to the manuscript:

"To determine the settings for the wind filters applied to two stations (GNS and OVS), we first defined the wind sectors corresponding to the previously identified emission hot spots. We then tested 14 combinations of parameters (wind direction and speed, and concentration variability over one hour) with wind speeds between 2 and 4 m/s and concentration standard deviations varying between 2 and 4 ppm for CO2, and between 4 and 10 ppb for CO and CH4. The optimal criteria (Table 2) were selected by comparing the GNS and OVS stations with the nearest stations not affected by local sources, namely AND and SAC-15m. We selected the combination of parameters (Table 2) that best eliminates contaminated values defined as hourly values being above a threshold (5 ppm for CO2, 25 ppb for CH4 and CO) compared to the nearest stations AND and SAC-15m respectively for GNS and OVS.

The impact of the wind sector filter differs between the two stations. In the case of the OVS site, we see on Figure 3 a strong impact on CO concentrations from the statistical filter, when it is applied to the entire data set. However, when applied to measurements that have already been filtered by wind sector, the statistical filter no longer makes large differences. This means that the wind filter has already eliminated the vast majority of spikes identified by the statistical method. A different result is obtained at the GNS station, where it can be seen, particularly for CO2 and CH4, that the impact of the statistical filter is similar whether it is applied to raw measurements or to measurements that have been filtered for wind sector beforehand. This last filter, based on wind, therefore has only a limited impact on the dataset. We hypothesize that this difference in behavior between the two stations, GNS and OVS, results from the distances between the sources and the analyzer's air inlet. In the case of OVS, the local source of contamination is close to the air intake and therefore clearly identified geographically. This means that the wind sector from which the contamination peaks originate is clearly identified. In the case of GNS, the origin of the contamination peaks is less precisely defined. As shown in Figure 5, a contamination source has been identified 3 kilometers away (water treatment plant), which is particularly active in terms of CH_4 emissions. In view of the results obtained, showing that the wind filter does not effectively filter out local contamination peaks, it appears that the GNS site is probably affected by several nearby sources of contamination and is therefore a difficult urban station to use as a representative of regional emissions."

4. Role of SAC100 as Background Site:

The manuscript uses SAC100 as a background reference for gradient calculations but also notes its altitude-dependent decoupling under stable conditions. Can the authors quantify how often SAC100 is truly representative of background air, maybe via PBLH diagnostics or footprint modeling? Presenting uncertainty

estimates for concentration gradients associated with this would improve the robustness of inversion-readiness claims.

With regard to the claim about the suitability of the gradients for inversion, it is clearly stated that the status of background site for SAC100 is "*depending on the wind direction*", and that it is not valid when, for example, the wind is coming from the north (12% of the time). We specify that AND and COU should be used as background in other wind directions: "*the background 'clean air' value of CO in the NE sector at the AND station, in the E sector at the COU station, and in the NE sector at the GNS station...*". Unfortunately, we do not have PBLH measurements for the Saclay site. In the study that we added to the paper on Paris offset trends (section 6), we specified the selection of Saclay data for use as a background site:

"*In order to obtain a time series that is as representative as possible of the regional background, we selected measurements from the Saclay tall tower (100 m high) when the wind was coming from the southwest (185°-285°), which is, incidentally, the prevailing wind direction, with a speed greater than or equal to 4 m/s. This dataset represents 33% of all measurements.* "

The wind speed criterion makes it possible to eliminate periods of high stability for which the definition of a background becomes indeed questionable.

5. Rationale for Analyzing Differences between the Measurement and the Temporal Average:

The authors calculate residuals by subtracting a 3-month rolling mean from the hourly time series and then examine the correlation between these residuals for CO_2 vs. CO and CH_4 vs. CO. However, this approach raises several concerns: 1) Since the main goal of this part of analysis is to understand emission-related co-variability, removing the seasonal cycle and long-term trends could obscure the very patterns of interest, particularly for species with strong source-seasonality like CO_2. It's unclear why this step is necessary. 2) More directly relevant would be to analyze co-variability between enhancements (e.g., ΔCO_2 or ΔCH_4 relative to a background site like SAC100), especially during downwind conditions. This would isolate the urban emission signal rather than mixing local and regional variability into a residual. This also aligns with how inversion systems interpret urban plume signals. 3) Arbitrary smoothing window? The use of a 3-month rolling average is not clearly justified. How sensitive are the correlations and slopes to this choice? Would the use of daily, weekly, or climatological baselines yield different results? I recommend that the authors either (i) shift the co-variability analysis to focus on signal–background enhancements, or (ii) more clearly justify and validate their use of detrended residuals.

1. The objective of this study is to characterize the signature of regional emissions for each measurement site. We believe it is necessary to isolate intra-seasonal variations, since long-term trends and seasonal cycles are strongly influenced by large-scale emissions. That said, trend analysis is extremely relevant, and we have therefore added a section to the manuscript describing, as suggested by this review, the long-term evolution of the offset (signal-background) of the different species measured (section 6). However, we believe it is important to study trends and detrended residuals separately.

2. Indeed, residuals can be calculated in multiple ways. Since our objective is to provide the chemical signature of each measurement site, whether located in the city center or on the outskirts, we have chosen a pragmatic approach, independent of uncertainties related to the choice of background depending on wind direction.

3. As suggested we conducted a sensitivity study on the smoothing window between 3 months and 1 week. Ultimately, the impact on the slopes is a maximum of 10% in winter and 13% in summer. This information has been added to the manuscript.

Minor Comments

Line 20: Specify how the reported baseline growth rates of CO2 (+2.34 ppm/year) and CH4 (+11.1 ppb/year) were computed (e.g., linear regression, seasonal detrending) :
Growth rates have been calculated with a liner regression as specified in the abstract

Line 61-69: The legend showing "Emissions totales" in Figure 1 was not explained in the main text or in the caption. Also, that information was not used in the analysis.
The background map showing anthropogenic CO2 emissions estimated by AIRPARIF has been modified in response to the review. There is now a clear indication of the unit of emissions represented.

Line 128-134: A brief description of ICOS protocols and classification is needed for readers to understand what is going on here.
We have developed the description of the calibration protocols : "*At CDS, AND, COU, GNS and OVS sites we use a common calibration scale made of three cylinders that are moved to each site periodically. These cylinders are filled with synthetic air, with concentrations ranging from 371 ppm to 504 ppm for CO2, 1646 to 2082 ppb for CH4, and 81 to 492 ppb for CO. In this article, ppm represents mole fraction of CO2 in dry air expressed in parts per million (or in parts per billion for ppb of CH4 and CO). The 3-cylinder common scale is sent to each site approximately once every 6 months. In between two calibrations, a reference gas is measured once a day in order to apply a one-point calibration and correct for short term drifts. The value of this reference tank is corrected by linear interpolation between two calibrations. A target gas is also analyzed once a day to evaluate the bias and repeatability of the time series (Figure S1). The way we use target gas information is explained in detail by Yver Kwok et al. 2015. Biases are defined as the differences from the assigned values of the target gases (Figure S1). The repeatability is expressed through the standard deviation of these daily injections of the target gases. The JUS site is equipped with its own calibration scale, consisting of 3 cylinders prepared in the same way as the common scale at the other stations. A calibration sequence is carried out every 1 to 2 months, and we do not use a reference gas*."

Line 155: Please clarify if CO data at OVS was used in the analysis due to the unsolved problem in its calibration.
The OVS CO measurements were used after correction for a bias of 14.1 ppb (lines 155-156). This is in fact the only case where the target gas measurements were used as a calibration correction.

Line 198-200: What does "the impact of spike filter" mean? Are you referring to the data filtered out or the data selected to use in the analysis?
The term "impact of spike filtering" refers to the difference between the initial data (without any filtering), and the spike-filtered data using the statistical filter. We have clarified this in the text.

Fig. 7: It's very hard to read. Consider changing it into a year-long monthly distribution plot.
For greater visibility, the figure was changed to monthly resolution as suggested.

L335-336: "outliers not detected by the filters" but removed as outliers here, which is ambiguous and confusing. More explanation is needed.
Indeed, the fact that the filters were not sufficient for the Gonesse station was not made sufficiently clear. We have detailed this point in the manuscript:
"*In the case of GNS, the origin of the contamination peaks is less easily identifiable than at OVS station. precisely defined. As shown in Figure 5, a contamination source has been identified 3 kilometers away (water treatment plant), which is particularly active in terms of CH4 emissions. In view of the results obtained, showing that the wind filter does not effectively filter out local contamination peaks, it appears that the GNS site is probably affected by several nearby sources of contamination and is therefore a difficult urban station to use as a representative of regional emissions*."

L350–355: Consider briefly explaining why "… amplitudes in CH4 diurnal cycles remain fairly stable across seasons and is likely driven by vertical mixing rather than emissions…" given that the residential sector showing strong seasonality (45% vs. 7%, Line 301-305).
Higher CH4 emissions from the residential sector in winter may explain a small part of the seasonal cycle, but its impact on the diurnal cycle appears to be low, which can be explained by the fact that the residential sector accounts for only 10% of annual emissions. The dominant sectors for CH4 (energy production, waste, agriculture) show little seasonal or diurnal variability. One sentence has been added in the text:
" The residential sector, which has the greatest seasonal and diurnal variability, accounts for only 10% of annual CH4 emissions, resulting in a relatively low impact in the atmospheric variability."

Line 496: "Bio-fuels may be responsible…" — please clarify if this is speculation or supported by inventory evidence/previous study.
This reference to biofuels has been removed.