Population exposure to outdoor NO_2, black carbon, particle mass, and number concentrations over Paris with multi-scale modelling down to the street scale

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Reply to reviewer 2

I have thoroughly reviewed this paper, which provides valuable insights by mapping the concentrations of various species down to the street scale. The study addresses an important aspect of air quality management and public health. However, after careful consideration, I regret to say that the manuscript does not meet the standard for publication in ACP at this time. Below, I outline my main concerns:

Calculation and representativeness for population exposure: The primary concern lies in the calculation of population exposure, as the PWC method used in this study is relatively simple and commonly found in previous studies. As it stands, the study does not offer a sufficiently novel approach.

Our reply: The novelty of our paper is threefold: 1. the consideration of BC and PN exposures, which are very rarely estimated with deterministic modelling, 2. the innovative modelling of residential exposures by coupling fine-scale urban mapping with detailed population data per building, 3. the estimation of exposure scaling factors between exposure at fine and regional scales for multi-pollutants.

This study focuses on population exposure to outdoor concentrations at residences, which is commonly used as a proxy for exposure in epidemiological studies (Hoek et al., 2024), or it is used as an input when estimating multi-environment exposure (Karl et al., 2019; Valari et al., 2020; Elessa Etuman et al., 2024). In epidemiological studies, exposure to outdoor concentrations at residences is often estimated using Land-Use Regression models (Ma et al., 2024), which are usually based on linear regressions using land-use predictor variables and data from fixed monitoring stations and passive sampling. Regional-scale models (chemical transport models with a spatial resolution often coarser than a few km^2) are sometimes used (Ostro et al., 2015; Adélaïde et al., 2021), leading to simulated fine PM concentrations much lower than those simulated using LUR models (Lequy et al., 2022). Their use is limited, because they are not able to represent the urban heterogeneities, e.g. gradient between street and background concentrations. Multi-scale models, i.e. a combination of regional and local-scale models (Kwak et al., 2015; Lee and Kwak, 2020; Park et al., 2021; Lugon et al., 2022; Lin et al., 2023; Wang et al., 2023; Strömberg et al., 2023), do represent urban heterogeneities, but they are often not able to represent the PM composition and the UFP, or their application is limited to a city district.

This multi-scale modelling of BC and ultra-fine particle is very novel, as it allows modelling down to the street scale over a whole city. Furthermore, using detailed population data per building, an indicator is presented to assess how much regional modelling underestimates population exposure for different pollutants. It varies depending on the pollutants and in particular on their urban variability. To our knowledge, this kind of estimation has never been done before.

The introduction has been rewritten using the sentences above to precise that exposure to outdoor concentrations at residences is studied and to better point out the novelty of this work.

Given that the key objective of this study is to assess the impact of spatial heterogeneities on population exposure from mobile emissions, a more advanced calculation of the PWC (or ESF) is necessary. For example, incorporating a spatiotemporally-weighted approach that accounts for the floating population could significantly enhance the analysis. Moreover, the current calculation of population exposure is limited to a specific temporal range, making it less representative of the broader situation in Paris. Expanding this temporal scope would provide a more comprehensive and accurate representation. Furthermore, returning to the basics, the street-level concentration shown in Figure 9 essentially reflects the PWC, making the conversion to a coarse resolution using Equation (1) less meaningful.

Our reply: Here, we do not aim to assess the impact of spatial heterogeneities on population exposure from mobile emissions, but from all sources. This is now specified in the introduction: "The coupled systems represent concentrations from the regional down to the street scales, taking into account all emission sources and secondary particle formation at all scales consistently (Lugon et al., 2022). "

The coupling CHIMERE/MUNICH allows to represent primary and secondary pollutant concentrations from the regional to the street scales with an hourly temporal resolution. Compared to the widely used LUR approach, the approach proposed here has the advantage of providing hourly concentrations, allowing short-term effects of pollution on health to be studied.

Compared to other deterministic approaches, it has the advantage of providing fine spatial resolution for multi-pollutants, including BC and UFP. As explained before, accounting for a floating population, i.e. multi-environment exposure, is completely out of the scope of the paper. The calculation proposed here for PWC is very advanced. Using deterministic modelling, studies often considers that exposure at residences is the same that exposure to regional background concentrations (Ostro et al., 2015; Adélaïde et al., 2021; Lequy et al., 2022), while the recent study of Lugon et al. (2022) assigns all the population to the major streets, without considering that people may also live in buildings that are not on the main streets. Here, we account for the spatial distribution of the population per building, in each street, providing much greater details than previous studies.

Although, as the reviewer mentioned, our current analysis is limited to a specific time period (summer time), a key objective is to present an improved method for assessing population exposure to BC and PN, and we have indicated plans for future evaluations covering other seasons, as well as the potential application of this methodology to other cities.

Lack of validation in modeling performance: Although the study includes some analysis in Tables 3 and 4, the comparisons are spatially limited. Model simulations should be compared spatially and temporally with observations to sufficiently (more intensively) verify the modeling performance. Presently, results are provided for only a few selected points without comprehensive statistical analysis, which diminishes the reliability of the findings. Including time-series analysis for each monitoring site, along with statistical metrics such as mean bias, RMSE, correlation coefficient, and index of agreement, would greatly improve the robustness of the result. Furthermore, there is insufficient evidence to demonstrate that simulations with the REF inventory outperform those with EMEP. In Figure 7, simulations of PM5 and PN using EMEP inventory appear to be more accurate, especially considering the following: i) Excluding the high overestimation of EMEP in the first bin of Figure 6 and ii) The Uncertainty is introduced by applying a mean conversion factor derived from one point to all points. Thus, if EMEP is applied in Figure 8, it seems likely that the performance of PM2.5, eBC, and NO2 would surpass that of the REF inventory.

Our reply: The model was evaluated at many background and traffic stations, using data from the monitoring stations from the air-quality agency AIRPARIF, as well as data from two field campaigns (ACROSS and sTREEt) as detailed in section 2.4. The model was compared to measurement at each station, and the comparisons were evaluated using many detailed statistical indicators with performance criteria from the literature, as detailed in Tables 2 and 3. Only the average indicators over the stations were shown in the main document, as showing the detailed evaluation per station does not provide any additional useful information. We have attached the tables of the statistical indicators along with the time series at each station in a Supplementary document.

In Figure 7, the eBC and PN concentrations are largely over-estimated with the EMEP emission inventory. In the case of $PM_{2.5}$, there is not a substantial difference between the REF and EMEP. For PN, EMEP tends to overestimate concentrations, particularly showing significant errors in the first bin, as the reviewer mentioned. At PRG, the second size section is also over-estimated using EMEP emissions. The reviewer mentioned the “Uncertainty introduced by applying a mean conversion factor derived from one point to all points to estimate eBC”. This is done independently of the emission inventory, and as recommended by Savadkoohi et al. (2024). In Figure 8, which aims at comparing regional and street scale concentrations, the

EMEP emission inventory can not be used because we can not use it at street scale as we do not downscale it further than 1 km x 1 km spatial resolution.

Lacks explanation of methodology: Given that the key aim of this study is to develop a method for simulating multiple pollutants down to the street scale, the manuscript would benefit from a more detailed explanation of the street-level model. The current manuscript focuses too much on emission data. This additional clarity would help readers better understand the approach and its applicability.

Our reply: The section on emission data was shortened and details were put in the Appendix. The WRF-CHIMERE/MUNICH/SSH-aerosol chain used in this study is the same as that employed by Maison et al. (2024). The street level model MUNICH has been thoroughly described in a lot of papers (some are mentioned in the introduction). The paper of Kim et al. (2022) is cited in section 2.1, because it describes the last official version of MUNICH. This study emphasizes the modelling of BC and PN, which requires careful processing of emissions, as well as comparisons to observations. In the model description of section 2.1, the models are briefly described as well as the differences with the official versions from which they were derived.

For clarity, the sentences mentioning the time of the study and the reference to the paper of Maison et al. (2024) are placed at the beginning of section 2.1: "Simulations are performed with WRF-CHIMERE/MUNICH/SSH-aerosol from 01 UTC on 1 June 2022 to 23 UTC on 31 July 2022, corresponding to a period with specific PN and BC measurements performed over Paris, as detailed in section 2.4. The setting is the same as in Maison et al. (2024), and it is summarized here."

Language quality: The English throughout the manuscript requires improvements, including correction of grammatical errors and typos. Many sentences are overly lengthy and would benefit from restructuring for better readability.

Our reply: We improved the quality of the language by carefully reviewing the manuscript.

Specific comments:

Figure 5 and Lines 185-194: Clearly define the following terms: i) non-traffic emission, ii) Exhaust emission, iii) non-exhaust emission, iv) other-traffic emission, and v) non-road traffic emission, mentioned in the manuscript. Based on the classification, it is unclear how aviation emissions (lines 185 – 194) are categorized under the non-traffic emissions.

Our reply: We have added new sentences to define the terms clearly in the revised manuscript.

"The distribution between traffic (exhaust vehicular road, non-exhaust vehicular road, non-road/other traffic) and non-traffic emissions for NOx, BC, PM, and PN over Greater Paris is shown in Figure 3, and over Paris in Figure C1. Non-traffic emissions refer to all emissions excluding traffic emissions (exhaust vehicular road emissions, non-exhaust vehicular road emissions, non-road/other traffic emissions). Exhaust emissions are produced from the combustion processes associated with road traffic emissions, while non-exhaust emissions include road-traffic emissions resulting from road wear, tire wear, and brake wear. Other-traffic emissions encompass emissions from ships and aircraft."

Lines 241-242 and Table 2: The size distribution described in Table 2 is not clearly explained, making it difficult to understand.

Our reply: The size distribution in Table 2 refers to the ratio between $PM_{2.5}$ and PM_{10} in the emissions. The following sentence has been added in the section that details the sensitivity study: "In the emission inventory, the non-exhaust emissions of particles are provided for PM_{10}. Emissions of $PM_{2.5}$ and PM_1 are estimated using a $PM_1/PM_{2.5}$ ratio and a $PM_{2.5}/PM_{10}$ ratio."

Captions: Review and correct the captions of Figures 4 and B5.

Our reply: The label was modified to "Regional (left panels) and street (right panels) traffic emissions of NO_2 [(a) and (b)] and BC [(c) and (d)] averaged from June to July 2022."

Section 2.2.5 Title: Clarify the title of Section 2.2.5: Emission sources of NOx (?), BC, PM, and PN.

Our reply: The emission sources for NOx were indeed included in our analysis, and we understand the importance of accurately representing this in the section title. We have revised the section's title to "Emission sources of NOx, BC, PM, and PN".

Figure order: The figures are not mentioned in the correct order. In the manuscript, Figures B3, B1, B2, B5, and B6 are referenced out of order. Figure B4 is not mentioned.

Our reply: Thank you for your observation regarding the figure order. We have revised the manuscript to reference the figures in the correct order as they appear. Figure B4 has been removed, as it was not referenced in the manuscript.

Methodology placement: The methodology for population exposure would be better placed in Section 2 (i.e., Section 2.4).

Our reply: As suggested, the methodology of population exposure was moved to section 2.

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