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 1

Overall Evaluation:

The study presents a valuable approach by using a coupled WRF-CHIMERE/MUNICH/SSH-aerosol model to simulate pollutant concentrations such as NO2, black carbon (BC), PM2.5, and particle number (PN) at the street level, and evaluating population exposure in the Greater Paris region. The topic is timely and important, especially given the underestimation of population exposure to pollutants like NO2, BC, and PN when only using regional-scale models. The study includes an impressive range of input data and models, and the results are potentially impactful. However, there are some significant issues related to the structure, clarity, and depth of analysis that need to be addressed to improve the overall quality of the paper.

Major Comments:

1. Imbalance Between Technical Details and Discussion:

While the technical details are thorough, there is insufficient analysis and discussion of the results. The paper would benefit from a deeper exploration of the trade-offs between traditional regional-scale models and street-level models. Specifically, *discuss how much computational resources are required for street-level models and how much additional accuracy is gained in comparison.*

Our reply: Sentences were added in Section 2.1 to further discuss the trade-offs between traditional regional-scale models and street-level models in terms of CPU time: “Using a one-way coupling approach, the regional-scale and local-scale simulations are performed sequentially. For the regional scale, the two-month simulation using WRF-CHIMERE models requires approximately 11520 hours×processors. The local scale simulations are less expensive, and the two-month simulation with the MUNICH model requires around 7680 hours×processors to simulate the street concentrations in the Parisian street-network composed of 4655 streets.

The advantage of the coupled system is now better emphasized 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). "

and it is also better emphasized in the conclusion: "The regional-scale simulation provides a comprehensive representation of urban background concentrations, but lacks the ability to estimate fine-scale concentrations. Conversely, the street-level simulation adopts a higher spatial resolution and provides more accurate concentration estimates, which are critical for assessing population exposure. The additional computational resources required for street-scale simulation are balanced by the improved accuracy in representing spatial variability, which is essential for effective urban air quality management.

Additionally, in order to provide a more precise analysis and validation of the model results, we have included tables of the statistical indicators at each measurement station in the Supplementary document.

2. Introduction Structure:

The introduction lacks a clear, logical flow and does not effectively highlight the key research gap. It would be helpful to reorganize the introduction to show a more coherent development of the research problem and objectives, making it easier for readers to understand the motivation behind the study.

Our reply: The introduction has been revised to improve its clarity and logical flow, effectively highlighting the key research gap. In particular, we emphasized the limitations and research gap in previous studies regarding population exposure compared to this study. The introduction is now structured in five main parts: importance to characterize BC and UFP concentrations, difficulties encountered in modelling BC and UFP, limitations in estimation of population exposure to outdoor concentrations at residences using deterministic modelling, limitations in urban modelling for multi-pollutants, and plan of the paper.

"In metropolises, characterised by densely populated and extensively developed areas, air pollution remains a major concern due to the presence of numerous emission sources, such as traffic, energy consumption, solvents, industrial activities. Traffic emissions receive particular attention because of their influence on local concentrations, with an impact of both exhaust and non-exhaust emissions (Fu et al., 2020; Jereb et al., 2021; Holnicki et al., 2021; Sarica et al., 2023). Environmental regulations aim to reduce key air pollutant concentrations, such as NO_2, O_3, and fine particulate matter ($PM_{2.5}$). Although a large part of the health impacts are attributed to particles (Southerland et al., 2022), the health effects associated with different particle compounds and different particle size can vary considerably (Park et al., 2018; WHO, 2021; Haddad et al., 2024). In particular, black carbon (BC) and ultra-fine particles (diameter lower than 0.1 μm) are considered "priority" emerging pollutants (WHO, 2021; Goobie et al., 2024), as stated in the European air-quality directives promulgated in October 2024. Long-term exposure to ultra-fine particles is associated with increased mortality (Li et al., 2023b), while BC has been linked to adverse health effects, especially in urban areas (Lequy et al., 2021; Bouma et al., 2023; Kamińska et al., 2023). Whereas fine particles are best characterized by their mass concentrations ($PM_{2.5}$), the mass of UFP is low compared to that of fine particles. Hence, UFPs are best characterized by their particle number (PN) concentrations (Kwon et al., 2020; Trechera et al., 2023), contributing to about 80-90% of the PN concentrations over urban areas (Dall'Osto et al., 2013; Abbou et al., 2024).

Although modelling is often use to assess the effect of emissions and policies to improve air quality in cities (Mao et al., 2005; Yuan et al., 2014; Kuklinska et al., 2015; Selmi et al., 2016; Andre et al., 2020; Lugon et al., 2022), assessments on BC and UFP concentrations are not frequently evaluated, because they are not regulated, nor measured routinely in cities and difficult to model. Difficulties to model BC are partly linked to differences between elemental carbon and black carbon (Savadkoohi et al., 2023), contributing to large model/measurement discrepancies (Lugon et al., 2021b). However, recommendations for assessing BC concentrations were recently provided by Savadkoohi et al. (2024). The PN concentrations are even more difficult to model, because of the lack of emission inventories and the rapid transformations of the ultra-fine particles involved (Kukkonen et al., 2016). The difficulties to model BC and PN might also partly be linked to the strong influence of traffic emissions on their concentrations (Andre et al., 2020; Jia et al., 2021; Lugon et al., 2022; Li et al., 2023a; Trechera et al., 2023). Traffic emissions are highly spatially and temporally variable in cities, and their variability is not easily reproduced in emission inventories. Those are usually built using either top-down or bottom-up approaches (Guevara et al., 2016). Bottom-up approaches use detailed spatial and temporal information for each activity sector, e.g., the number of vehicles for traffic emissions, while top-down approaches use information defined at larger scales (regional or national), which are spatialized using specific data, such as population data. Significant discrepancies may exist between emission inventories using these two approaches (Guevara et al., 2016; Lopez-Aparicio et al., 2017), especially for traffic emissions (Lopez-Aparicio et al., 2017) and non-exhaust emissions from tire, brake and road wear (Piscitello et al., 2021; Tomar et al., 2022). Emission inventories for UFP only exist for top-down inventories (Kulmala et al., 2011; Zhong et al., 2023). Sartelet et al. (2022) recently provided a methodology to estimate PN emissions from any emission inventories of PM, making it possible to use either bottom-up or top-down emission inventories.

Population exposure to outdoor concentrations at residences 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 residence 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. gradients 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., 2023b; 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. To represent an entire city, chemical-transport models are often coupled with a simple representation of local dispersion (Hood et al., 2018; Karl et al., 2019; Lugon et al., 2022; Maison et al., 2024) or with subgrid statistical approaches (Valari and Menut, 2010; Squarcioni et al., 2024). However, only a few studies model BC (Lugon et al., 2021b) and PN (Zhong et al., 2023; Ketzel et al., 2021) concentrations down to the street scale. To model PN, the main difficulty lies in the evaluation of atmospheric transformations (Kukkonen et al., 2016; Strömberg et al., 2023), and there is to our knowledge no multi-scale model currently available to represent PN over a whole city from the urban background down to the street scale taking into account aerosol dynamics. To simulate gas and particle concentrations over cities from the regional down to the local scale taking into account chemistry and aerosol dynamics, the chemical module SSH-aerosol (Sartelet et al., 2020) has been coupled with air quality models at the street and regional scales: the street-network Model of Urban Network Intersecting Canyons and Highways (MUNICH) (Kim et al., 2018, 2022) and the regional-scale models Polair3D (Lugon et al., 2021a, 2022; Sarica et al., 2023; Sartelet et al., 2024) and CHIMERE (Maison et al., 2024; Squarcioni et al., 2024). The coupled multi-scale 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; Sartelet et al., 2024). Although extensive comparisons to observations were performed at the regional and local scales for NO_2, $PM_{2.5}$, and PM_{10} (Sartelet et al., 2018; Lugon et al., 2022; Kim et al., 2022; Sarica et al., 2023), urban multi-scale modelling evaluation of BC and PN at both regional and local scales is still missing."

3. Parallel Treatment of PN and Pollutants:

Particle number (PN) is a statistical measure, not a pollutant like BC or PM2.5. It would be clearer to first discuss pollutant concentrations and then evaluate particle characteristics through PN. Avoid listing PN alongside BC and PM2.5 in a parallel manner in the title and main text.

Our reply: PN is not a statistical measure, it corresponds to the particle number concentrations. It is a metric used to represent particles (Kwon et al., 2020; WHO, 2021), similarly to $PM_{2.5}$. $PM_{2.5}$ represents the mass of particles of diameter lower than 2.5 μm, and PN represents the number of particles. $PM_{2.5}$ is mostly influenced by fine particles of diameters in the range 0.1 to 2.5 μm, whereas PN is mostly influences by ultra-fine particles, of diameters lower than 0.1 μm. Hence, for clarity, the title was modified to "Population exposure to outdoor NO_2, black carbon, ultra-fine and fine particles".

4. Excessive Abbreviations:

The use of abbreviations is sometimes excessive, making the text difficult to follow. For instance, in Line 44, the abbreviation "PN" is introduced without proper context. It is recommended to reduce the use of abbreviations, particularly for terms like "particle number," to improve readability.

Our reply: The abbreviation for PN is commonly used in the literature, and now properly introduced in the introduction. In this paper, the abbreviation "PN" is consistently used throughout the discussion of the paper's results. To improve readability, a table summarizing the different abbreviations was added at the beginning of section 2.

Table 1. List of abbreviations

	Nomenclature
BC	Black carbon
eBC	Equivalent Black Carbon
PN	Particle number
LUR	Land-Use Regression
CTM	Chemical transport model
UFP	Ultra-fine particles
REF	Reference simulation
SEN	Sensitivity simulation
PWC	Population Weighted Concentration
ESF	Exposure Scaling Factor

5. Clarification on PN and Ultra-fine Particles:

The introduction mentions that ultrafine particles are best characterized by PN concentrations. If PN is being used in this study to represent ultrafine particles, ensure this connection is well-supported in the text. If the simulation does not focus on ultrafine particles, it would be better not to mention them, as they are challenging to model accurately.

Our reply: One main focus of this study is the modelling of ultra-fine particles, which are characterized by their number concentrations, because their mass is low but their number is high, as detailed in the reply number 3 to the reviewer's comments. The introduction has been rewritten to better explain this point: "Whereas fine particles are best characterized by their mass concentrations, the mass of UFP is low compared to that of fine particles. Hence, UFPs are best characterized by their particle number (PN) concentrations (Kwon et al., 2020; Trechera et al., 2023), contributing to about 80-90% of the PN concentrations over urban areas (Dall'Osto et al., 2013; Abbou et al., 2024)." Furthermore, the title has been changed to bring up the terms fine and ultra-fine particles.

6. Model Setup and Emissions Summary:

The description of the model setup and emissions data in Section 2 is too detailed and could be streamlined. Consider summarizing the key aspects in a table and moving the detailed descriptions to the supplementary information (SI) for clarity.

Our reply: The details about the speciation of traffic and non-traffic emissions were put in the Appendix B. To improve clarity, the order of the sections was modified, with the section on the "Sensitivity to non-exhaust emissions" presented after the section on "PN emissions". As suggested by reviewer 2, the description of the calculation of exposure was added to the "Material and methods" section.

7. Quantitative Differences in Section 3.3:

Instead of using qualitative terms like "higher" or "lower" in Section 3.3, it would be more informative to present the quantitative differences between the results to enhance the clarity of the comparisons.

Our reply: We have added the quantitative difference in section 3.3.

Now, it reads: "The impacts of the emission inventory are investigated over Greater Paris. In the EMEP simulation, the NO_2 concentrations are lower by about 15% along the roads and airport than those in the REF simulation, due to higher traffic emissions using the bottom-up inventory than the EMEP one (Figure C5). In contrast, NO_2 concentrations in areas excluding roads are higher, by about 16%, in the EMEP simulation. The concentrations of NO_2, eBC, $PM_{2.5}$, and PN in the EMEP simulation are 12%, 50%, 7%, 38% lower, respectively, for Paris compared to those in the REF simulation. The differences in the spatial distributions of eBC and $PM_{2.5}$ concentrations are similar to those of NO_2 concentrations, but the spatial differences are less pronounced than for NO_2. In the eastern region of Greater Paris and the extreme northwest part of the region, PN concentrations are lower using EMEP than the bottom-up inventory owing to lower emissions compared to other areas within the region."

8. Applicability to Other Cities:

This study focuses on street-level traffic emissions and population distribution in the Greater Paris region. It would be beneficial to discuss whether the findings and conclusions could be extended to other large cities, particularly those with different urban structures or traffic patterns.

Our reply: As the reviewer mentioned, while this study provides insights into street-level traffic emissions and population exposure specific to Greater Paris, its methodologies and findings could be adapted to other major cities by reflecting the different urban structures and regular conditions. We have added the following sentences in the conclusion part:

“Multi-scale simulations using bottom-up traffic emissions provide innovative and detailed spatially-resolved air-quality information in urban areas. In particular, the ESF may be used to refine the evaluation of population exposure in urban areas employing regional-scale models. The methodologies and findings could be adapted to other major cities, with detailed street-scale emission inventory and street characteristics. This could be done for example, by the continuation of the modelling with the MUNICH model in several cities (Sarica et al., 2023; Wang et al., 2023a; Cevolani et al., 2024). Further investigation is also needed to assess the concentrations and population exposure scaling factors for different seasons.”

Minor Comments:

1.Line 19: Clarify what is meant by "regional scale"—what specific area or distance does this term represent?

Our reply: This is now clarified in the introduction: " Regional-scale models (chemical transport models with a spatial resolution often coarser than a few km^2)"

2.Line 33: Reword the sentence to clarify that not all particle compounds impact health equally.

Our reply: We have modified the sentences to clarity.

Now, it reads : “Although a large part of the health impacts are attributed to particles (Southerland et al., 2022), the health effects associated with different particle compounds and different particle size can vary considerably (Park et al., 2018; WHO, 2021; Haddad et al., 2024)."

3.Line 34: The term "large health effect" is vague; it would be more effective to provide specific examples or references.

Our reply: We agree that the term ’large health effects’ is vague. To enhance clarity, we modified the sentence to specify the health impacts of BC and ultra-fine particles.

Now, it reads: “In particular, black carbon (BC) and ultra-fine particles (diameter lower than 0.1 μm) are considered "priority" emerging pollutants (WHO, 2021; Goobie et al., 2024), as stated in the European air-quality directives promulgated in October 2024. Long-term exposure to ultra-fine particles is associated with increased mortality (Li et al., 2023b), while BC has been linked to adverse health effects, especially in urban areas (Lequy et al., 2021; Bouma et al., 2023; Kamińska et al., 2023)."

4. Line 36: Include a reference or example of the ratio of ultrafine particles in terms of number concentration, such as "XX% of total number concentration is from ultrafine particles."

Our reply: The following sentence was added in the introduction "UFPs are best characterized by their particle number (PN) concentrations (Kwon et al., 2020; Trechera et al., 2023), contributing to about 80-90% of the PN concentrations over urban areas (Dall'Osto et al., 2013; Abbou et al., 2024)."

5. Line 44: I don't think influences of traffic on BC is rare. https://www.sciencedirect.com/science/article/pii/S0269749121014500 https://acp.copernicus.org/articles/23/6545/2023/acp-23-6545-2023.pdf

Our reply: The term 'rare' in the sentence means that research on BC remains much less extensive compared to gaseous pollutants and PM. The references you provided were also published recently, and we added them to the paper in the illustration of the influence of traffic on BC and PN. The sentence has been modified to "Although modelling is often use to assess the effect of emissions and policies to improve air quality in cities (Mao et al., 2005; Yuan et al., 2014; Kuklinska et al., 2015; Selmi et al., 2016; Andre et al., 2020; Lugon et al., 2022), assessment on BC and UFP concentrations are not frequently evaluated, because they are not regulated, not measured routinely in cities and difficult to model. "

6. Lines 46-53: The paragraph discussing BC estimation and adjustment methods should be moved to the Methods section for better flow.

Our reply: We have moved the paragraph on the harmonization factor to Section 2 under Measurements, to improve the flow and organization of the manuscript.

7. Line 111: Spell out the full name of "SSH-aerosol" and clarify its role in the study. Similarly, "CAMS" at Line 118 should be defined.

Our reply: The SSH-aerosol model integrates three modules: SCRAM (Size-Composition Resolved Aerosol Model), which addresses the dynamic evolution of aerosols; SOAP (Secondary Organic Aerosol Processor) for gas/particle partitioning of organic compounds; and H^2O (Hydrophobic/Hydrophilic Organics) focusing on the formation of condensable organic compounds. Additionally, CAMS stands for the Copernicus Atmosphere Monitoring Service. We have clarified these terms for better understanding.

Now, it reads : "The chemical scheme used is MELCHIOR2 modified to represent the formation of organic condensables as described in SSH-aerosol (Sartelet et al., 2020), which is used for aerosol dynamics (coagulation and condensation/evaporation). The SSH-aerosol model integrates three modules: SCRAM (Size-Composition Resolved Aerosol Model), which addresses the dynamic evolution of aerosols; SOAP (Secondary Organic Aerosol Processor) for gas/particle partitioning of organic compounds; and H^2O (Hydrophobic/Hydrophilic Organics) for the formation of condensable organic compounds."

8. Figure 3: To better highlight the changes in NO2 and BC, consider normalizing the data to make the differences clearer.

Our reply: The normalized data would not significantly change the observed emission patterns between weekdays and weekends 1. Moreover, normalized emissions may not effectively represent the quantitative differences in emissions between NO_2 and BC or between stations. Therefore, we have decided to keep the original figure in μg m^{-2} s^{-1}.

* normalized emissions = emissions / average emissions

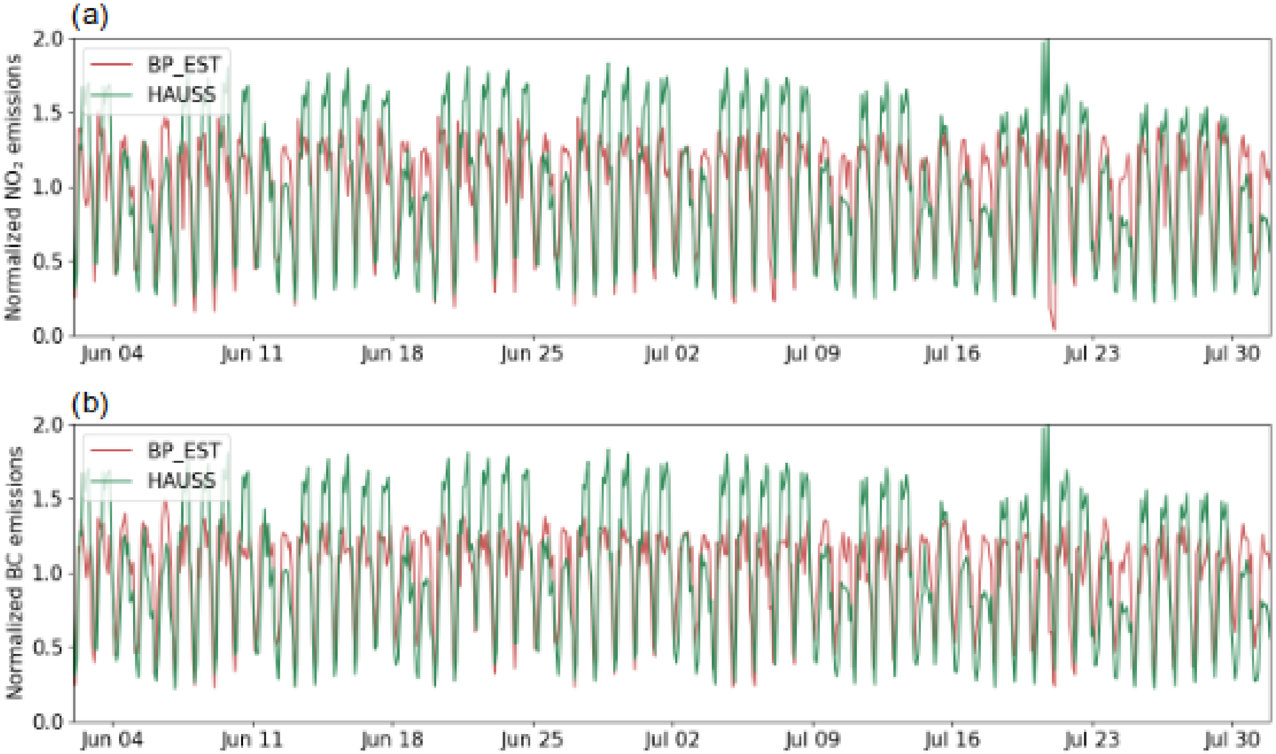

Figure 1. Time series for the normalized (a) NO_2 and (b) BC emissions at the HAUSS (city center) and BP_EST (heavy-traffic) stations

9. Line 113: Specify whether the WRF-CHIMERE/MUNICH coupling is online or offline.

Our reply: We already specify in the model description the coupling between CHIMERE and MUNICH: "The WRF-CHIMERE/SSH-aerosol model is one-way coupled to the street-network model MUNICH". We added details about the coupling between WRF and CHIMERE: "The CHIMERE model is coupled with the meteorological model Weather and Research Forecasting (WRF) (Powers et al., 2017), which was used to compute the meteorological fields needed in the simulation. Here, no feedback interactions are considered between concentrations and meteorological fields, with a one-way coupling approach."

10. Line 254: The formula presented here should be centered and numbered for clarity and consistency.

Our reply: We ensured the formula are centered and properly numbered for clarity and consistency in the revised manuscript.

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