Response to the reviewers on essd-2024-80 "Multi-year high time resolution measurements of fine PM at 13 sites of the French Operational Network (CARA program): Data processing and chemical composition"

We thank the reviewers for all the constructive comments. In the following, we respond to the reviewers using a black font for original review comments, green font for authors' responses, and blue font for changes in the revised version.

Please note that the line numbering corresponds here to those in the preprint initially submitted and not in the revised version.

For clarity, comments have been numbered.

Reviewer #1

General comments:

This study by Chebaicheb et al. present a unique dataset with multiannual (2016-2021) measurements of ACSM and AE33 collected at 13 urban sites in France including submicron (PM1) aerosol species, OA, NO3, NH4, SO4, Cl and eBC. Detailed description of the measurement instrument, data handling and data quality control are documented. This is of particular importance for data users. In addition, the authors conducted comprehensively analysis of the datasets, for example composition fractions, seasonal and diel cycles of each site are presented. Also, the dataset is used to evaluate the chemical transport model (CHIMERE) simulation. Overall, the topic is suitable for ESSD, and the manuscript is clear written. I would support for publication in Earth System Science Data after some corrections and clarifications.

We thank the reviewer for the valuable comments, which are addressed one by one hereafter.

1) About measurement uncertainty, since the authors trying to deliver an important dataset to the community, it's crucial to discuss about the uncertainty of each reported variable. Is there any limitation of the measurement that the data user should be aware?

We have added a paragraph in the main text about measurement uncertainties in section 2.4, which also aims to respond to comments #10 and #13: "Reconstructed PM_1 may overestimate measured $PM_{2.5}$ loadings mainly due to the respective measurement uncertainties of each technique used here. For $PM_{2.5}$, the FIDAS instrument has been demonstrated as equivalent to the EN12341 standard method with a maximum overall uncertainty of 25 % compared to this reference method according to EN16450 (Amodeo, 2024). It should also be stated that this instrument is sensitive to particles above 180 nm optical diameter only, which may result in even higher uncertainties for the estimation of the PM1 mass fraction. For eBC, a recent intercomparison between 23 AE33 devices (Cuesta-Mosquera et al., 2021) in the framework of the ACTRIS research infrastructure showed that the total mean deviation of the eBC concentrations at 880 nm for the 23 instruments was -2 % (range: -16 % to 7 %) before maintenance and -1 % (range: -14 % to 8 %) after maintenance, for soot measurements, emphasizing that the unit-to-unit variability was not significant. In our case, the post-processing of the datasets is the same for every site, therefore ensuring the comparability of the obtained concentration values. However, the main uncertainty in eBC concentrations lies in the various correction factors applied and not in the raw measurement itself. Considering the various approaches commonly used to transform absorption coefficients into eBC mass concentrations, and related propagation of errors, an overall uncertainty of up to ±50 % can be associated with eBC estimates (Savadkoohi et al., 2024). Eventually, the Q-ACSM has been shown to display reproducibility uncertainties of 9 % on NR-PM_1 measurement, with uncertainties of 15, 19, 28, and 36 % for NO_3, Org, SO_4, and NH_4, respectively (Crenn et al., 2015). The high uncertainties of SO_4 may be related to the RIE SO_4, especially since it was considered constant in the early years. Additional uncertainties are related to possible measurement artifacts associated with interferences due to the nitrate (and sulfate) signal (e.g., the Pieber effect on the CO_2^+ signal at m/z 44; Pieber et al., 2016). This artifact is explained by NO_3 (or SO_4)-induced reactions on the vaporizer and ionizer surfaces, producing CO_2 and therefore increasing the m/z 44 signal that is otherwise attributed to the organic aerosol. It can be quantified and evaluated over time by tracking the m/z 44 / NO_3 (m/z 30 / SO_4) ratios during the different calibrations performed with pure ammonium nitrate (ammonium sulfate) solutions. During the ACSM intercomparison at ACMCC in 2016 (Freney et al., 2019), the m/z 44 / NO_3 ratio was determined to vary between 0.01 and 0.26 for 15 instruments, and the m/z 30 / SO4 ratio between 0.01 and 0.173. These were checked for each instrument in this study using calibration data and the results obtained fell within these ranges thus no correction was applied. The overestimation of PM_1 could also be linked to a change in the chemical composition of organic aerosols when this fraction dominates (e.g. Nault et al., 2023, Xu et al., 2018), since for organics the RIE is considered constant (1.4 by default) and these species are not considered in the Middlebrook correction (Middlebrook et al., 2012). Finally, other uncertainties can be related to size selection. It should be noted that the ACSM aerodynamic lens system is considered to be fully efficient for particles from 40 nm up to 600 nm (Liu et al., 2007), while recent studies are suggesting collection size ranges that might be considered as instrument-specific (Poulain et al., 2020)."

2) Data availability: according to ESSD policy, I think there should be an individual section describing the dataset structure, unit, user guide, etc.

Indeed, we have a "Data availability" paragraph after the conclusions where we present the link to the data and in this link, we have a "readme" file that explains the data (the different variables, temporal resolution, detection limits, and geographical information in a table. We have added the unit in the revised version. Technical information and data validation are presented in the article (which, in some cases, also presents the data user guide).

3) The manuscript gives me the feeling that it is describing data (composition fractions, seasonal and diel cycles). While it seems lack of scientific contributions except for the data itself. Maybe this is fine for ESSD. I would suggest some more in depth discussion (for example, next point).

We thank the reviewer for this comment. Indeed, the aim of the ESSD journal is to describe the data, but we have taken this comment into consideration when revising section 3.3.

4) General suggestion about Section 3.3: I would suggest extend more analysis in this section by utilizing the dataset to evaluate model simulation and provide more insights to the model development. For example, it would be interesting to provide some maps

about spatial distribution, and focus on evaluation of CTM instantaneous simulation of hourly mass concentration?

We have developed this section, as also proposed by Reviewer #2, in a new Section 4.

"4. Comparison between observations and the CHIMERE Chemical Transport Model

Measurements of PM chemical composition are a valuable tool for validating atmospheric CTMs, particularly for assessing their accuracy and reliability. In particular, observations and model outputs are complementary to track complex atmospheric sources and processes, including chemical transformations leading to secondary PM formation. Comparing chemically-speciated observations with CTM model results enables discrepancies to be identified and could provide clues on model improvement. In addition, near-real-time observations allow gauging a model ability to represent the temporal and spatial distributions of atmospheric pollutants, which is essential for forecasting air quality and assessing environmental policies and scenarios. The continuous observations provided by the CARA program are of great importance for the continuous improvement of 3D air quality models, notably CHIMERE, leading to more accurate forecasts and a better understanding of atmospheric processes.

4.1 Model description

In order to exemplify the comparison of our database with CTM's outputs, 3D simulations were performed with the CHIMERE version of Wang et al. (2024) which is based on a coupling between CHIMERE (Menut et al., 2021) and SSH-aerosol v1.3 aerosol model (Sartelet et al., 2020). The Secondary Organic Aerosol (SOA) mechanism of Wang et al. (2024) was used. This mechanism was obtained by using the GENOA (*GENerator of reduced Organic Aerosol*) v2.0 algorithm (Wang et al., 2022, 2023) to reduce the SOA mechanisms for monoterpenes and sesquiterpenes from the Master Chemical Mechanism (Saunders et al., 2003) coupled with PRAM (accounting for SOA formation from monoterpenes by auto-oxidation) (Roldin et al., 2019). Following Wang (2023), the hydrophilic/hydrophobic organics (Chrit et al., 2017) mechanism was used for other precursors. Primary organic aerosols are treated as semivolatile organic compounds that partition as a function of environmental conditions and can undergo ageing (Couvidat and Bessagnet 2021).

One important feature of SSH-aerosol consists in the computation of gas-particle partitioning with the thermodynamic module ISORROPIA (Nenes et al., 1998) and SOAP (*Secondary Organic Aerosol Processor*, Couvidat and Sartelet, 2015) models for inorganic and organic aerosols, respectively. The latter accounts for the condensation of semivolatile organic compounds onto the organic and aqueous phases of particles as well as the effect on partitioning of interactions between organic and inorganic compounds based on their molecular structure. Thermodynamic equilibrium was assumed for gas-particle partitioning.

Meteorological data were obtained from the operational analysis of the Integrated Forecasting System (IFS) model of the European Centre for Medium-Range Weather Forecasts (ECMWF) (Flentje et al., 2021). Boundary conditions were taken from CAMS CIFS (IFS coupled to a tropospheric chemistry scheme) global model simulations (Flentje et al., 2021) for chemical species. Anthropogenic emissions of gases and particles were taken from the CAMS-REG-AP inventory at a 0.05°x0.1° grid resolution (version v5.1_REF2.1) (Kuenen et al., 2022).

4.2 Comparison results

CHIMERE model results for the year 2018, with a spatial resolution of 7 km over France, were used to compare with PM_1 observations at nine of the sites where data were available (excluding BPEst, Paris Les Halles, Rennes, and Strasbourg). The time series of observed and modeled concentrations are shown in the supporting material (Figure S10). Figure 6 summarizes results from the comparison between observations and simulations, typically showing good agreement. Loadings for inorganics (NO_3, SO_4, NH_4, and Cl) and eBC are fairly well captured by the model across all sites, with some exceptions. In particular, at the Marseille-Longchamp site, SO_4, NO_3, NH_4, and eBC are consistently underestimated by the model (33, 41, 45, and 65 %, respectively). This discrepancy could be due to the low resolution of the model grid (0.0625° x 0.125°) that may not be sufficient to capture local meteorology or sources, or more broadly a potential underestimation of emissions in the Southeastern region of France. Several sites also present an underestimation of SO_4 (Metz, SIRTA, Talence) by around 35-39 %. In contrast, NO_3 is strongly overestimated by the model (57 %) in the north of France (ATOLL). Organics, on the other hand, are consistently underestimated by the model at all sites by a factor of 2-3. Since eBC is well represented as discussed above, this leads to a low OA/eBC ratio, suggesting an underestimation of secondary organic aerosols in the model. Other recent studies also reported underestimations of OA at 11 European sites, focusing on winter 2009 (Ciarelli et al., 2016). In the present study, OA yields a strong underestimation particularly in the warmer months (60 % vs. 41 % for the colder months).

Figure 7 displays the diel profiles of each species, comparable with Figure 5, for the winter and summer of 2018 (spring and autumn profiles can be found in the SI, Figure S11). In general, the species exhibit relatively consistent model performance between winter and summer, although there is an underestimation by the model for the latter. For NO_3, the concentrations observed during wintertime are relatively stable throughout the day, whereas the model shows a strong daytime decrease due to the modeled volatilization of ammonium nitrate. During summertime, an enhancement of NO_3 in the early morning is captured by both observations and model, however as a smooth nighttime increase/decrease for the former, and a sharp peak in the latter. A similar pattern is observed for NH_4. For SO_4, the diel profile is quite constant for both observations and simulations in summer. In winter, the slight increase of SO_4 during the day is not captured by the model, which instead shows a low peak at night. For eBC, both observations and model simulations show two peaks during rush hours. In winter, the night peak is more pronounced in the model, but nonetheless they display comparable levels, in contrast to summertime, when the model tends to underestimate the concentrations. These differences in daily eBC profiles may be attributed to meteorological conditions or issues in the seasonal temporality of emissions. Finally for OA, as discussed above, the model largely underestimates observations in summer. Generally, the behavior is fairly well represented, however the wintertime nighttime enhancement is larger than observations, similar to eBC.

Figure 8 presents some statistical parameters (mean bias, normalized *Root Mean Square Error* (RMSE), and correlation coefficient r) calculated from the daily means for each chemical species across the nine urban sites in France. Overall, the correlations between observations and model results show good agreement, with correlation coefficients (r) ranging between 0.6 and 0.8, which is consistent with the literature (Couvidat et al., 2018, Cholakian et al. 2018). The mean bias and normalized RMSE confirm the model robustness. Mean bias is nearly negligible for SO_4, NO_3, NH_4, Cl, and eBC, and approximately -2 µg m^{-3} for OA, up to -4 µg m^{-3} for the Marseille Longchamp site. RMSE exhibits a slightly more scattered distribution, generally ranging between 0.5 and 2 µg m^{-3}.

These comparisons between PM_1 observations and model simulations reveal underestimations or overestimations by the model for each species. However, it remains challenging to pinpoint the exact reasons for these discrepancies, though hypotheses can be made. Generally, there is good agreement for SO_4. On the other hand, significant peaks of modeled NO_3 and NH_4 are observed, particularly in November and December at northern France stations, which may be explained by an overestimation of NH_3 emissions during this period in the model (Couvidat et al., 2018). For eBC, the results vary from one station to another, which may be linked to issues with the spatial distribution of emissions, which are not sufficiently accurate. OA is consistently underestimated across all stations. Further speciation of OA could provide more insights in this regard, which will be discussed in a forthcoming article on OA sources. Ultimately, conducting further simulations over other periods could help improve the model.

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Specific comments:

5) Abstract L37: 43-60%, mass or volume? It's good to clarify.

Mass, we have added it in the abstract: "…. Overall, OA dominates PM_1 at each site (43-60 % of the total mass), …"

6) Abstract L46: please expand CHIMERE

CHIMERE is not an acronym; it is the name of the model.

7) L75: About BC effects on climate (Jacobson et al., 2001), I believe there are many recent research results here.

Indeed, we have removed this reference and replaced it with a more recent one (Forster et al., 2023) in the main text and the references.

Instrument	type	Advantages	Disadvantages
AMS	cToF-AMS or HR-ToF-AMS	- field campaign - size distribution - high-time (~2-5 min) and mass resolution (HR-ToF-AMS)	- require more monitoring and technical maintenance
ACSM	-Q-ACSM	- long-term measurements - less maintenance - time resolution of ~30 min	- no size distribution - unit mass resolution
	-ToF-ACSM	- long term measurements - time resolution of about 10min - higher mass resolution ($m/\Delta m \approx 300$)	- no size distribution - data treatment procedures relatively complex

1) L89: please specify what's CARA program.

Added: "... the CARA program (Chemical characterization of particulate matter, set up in 2008 by the French reference laboratory for air quality monitoring) …"

2) Section 2.2.1: here you mentioned ACSM, Q-ACSM, ToF-ACSM, AMS, I'm not familiar with them. In my opinion, it would be helpful for readers to provide a table to summarize some key aspects (advantages/disadvantages, uncertainties, etc.).

We have added this table in the SI:

Table S2: Key aspects of AMS/ACSM instruments

In the main text, we have added this sentence in L132: "More information about these instruments is presented in Table S2."

3) L244: MAC_ACTRIS 7.5 m2/g, I think you refer to BC. In my opinion, this value is with high uncertainty that can vary from about 4 to >10, and it could have huge effects on your measured eBC values. I would suggest some more discussions about it.

Absolutely, and we have described this in section 2.3.2, in accordance with recent procedures as explained in Savadkoohi et al. (2024). First, the harmonization factor was introduced by ACTRIS to standardize the calculation of absorption coefficients using Aethalometers and depending on the filter tape used. Moreover, the MAC site- and season-dependence strongly affects the determination of eBC mass concentrations. The average MAC used here is considered as representative of European background conditions.

We have reworded this paragraph in the revised version: "eBC concentrations are obtained at a wavelength of 880 nm, where it is less prone to artifacts caused by other light-absorbing compounds such as dust (notably iron oxides) and some organic compounds (termed brown carbon, BrC, which absorb light at shorter wavelengths in the UV spectrum). In ambient air, the MAC value varies from site to site and from season to season, which affects the quantification of eBC mass concentrations. The harmonization factor was introduced by ACTRIS to standardize the calculation of absorption coefficients, depending on the filter tape used. At 880 nm, the MAC_{ACTRIS} factor used here is equivalent to 7.5 $m^2 g^{-1}$, also in good agreement with results previously obtained by Zanatta et al. (2016). It should be noted nonetheless that the application of the harmonization factor and the subsequent recalculation of eBC using a default and constant MAC value result in a reduction of about 40 % for eBC levels compared to the instrument raw outputs widely used in previous pan-European studies (such as Chen et al., 2022)."

4) L250: could you justify here why you decide the acceptable AAE 0.7 to 3.0? What's your explanation of measured values outside this range?

Particles emitted as a result of fossil fuel combustion have an α coefficient generally between 0.8 and 1.1. Higher values of α reflect a strong absorption of particles in the UV range and are observed for particles resulting from biomass combustion in particular, with α values typically up to 2.5. So, in ambient air, a range of values from 0.7 to 3 can be considered as realistic, and values measured outside this range can be considered outliers.

To address your comment, L256 has been changed into: “Lower and upper acceptable AAE values of 0.7 and 3.0 are arbitrarily considered here …”

5) L264: what do you mean NR-PM1? Non-Refractory?

Exactly, we have defined it now in the text L125: “… non-refractory submicron aerosols NR-PM_1 …”

6) Figure S1: sometimes PM1>PM2.5, could you please clarify and elaborate?

We have provided some explanations in the reply to comment #1.

7) L290: Wang, (2023) -> Wang (2023)

Done in the revised version.

8) L338-339: the measurements are conducted in daytime only?

We have corrected as follows: “Moreover, an increased mixing layer height over the Paris city center, due to the urban heat island effect which may dilute the aerosol content in a wider volume during daytime, should also be considered when comparing concentrations from inner and suburban sites within such a megapolis (e.g., Dupont et al., 2016).”

9) L351: it’s not clear what are the two sides you are comparing (22-30 % vs 9-20 %)?

Here, we are comparing the average contributions at northern sites vs southern sites: “… NO_3 contributions are more pronounced at northern sites (22-30 %, vs 9-20 % at southern sites), …”

10) Section 3.3: -> Comparison between observations and CHIMERE Chemical Transport Model

The title has been changed and section 3.3 has become section 4, following comment #5 from Reviewer 2:

“4. Comparison between observations and the CHIMERE Chemical Transport Model”

11) L468: I guess the spatial resolution of emission inventories you input into CHIMERE is even coarser than 7 km.

The emissions are taken from the CAMS-REG-AP inventory at a 0.05°x0.1° grid resolution (Kuenen et al., 2022) and are slightly more resolute than the simulations (0.0625°x0.125°).

12) L482: OA is underestimated by a factor of 2-3 at all sites, while eBC is more or less ok (a factor of < 1.5). Could you elaborate more about this? Or could you suggest something about OA/BC ratio?

We have developed as follows: “Organics, on the other hand, are consistently underestimated by the model at all sites by a factor of 2-3. Since eBC is well represented as discussed above, this leads to low modeled OA/eBC ratios (2.7-5.2, *vs* 3.9-8.8 for observed OA/eBC ratios), suggesting an underestimation of secondary organic aerosols in the model.”

13) General suggestion about Section 3.3: I would suggest extend more analysis in this section by utilizing the dataset to evaluate model simulation and provide more insights to the model development. For example, it would be interesting to provide some maps about spatial distribution, and focus on evaluation of CTM instantaneous simulation of hourly mass concentration?

Please refer to comment #4

Data availability: according to ESSD policy, I think there should be an individual section describing the dataset structure, unit, user guide, etc.

Please refer to comment #2

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