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 #2

The current work describes a new dataset of high-resolution PM1 measurements from the CARA program. The methodology is clearly described and the manuscript is well written. Measuring the chemical composition of PM is important in air pollution science, being a vital step towards the source allocation of air pollutants. I think the article is suitable for publishing in ESSD, but after the below comments are addressed by the authors.

We appreciate the reviewer's insightful comments, which are addressed one by one hereafter.

Section 2.5

1) Since the SOA mechanism of Wang et al. (2024) (using GENEO and SSH-aerosol) is described in Wang et al. (2024) as using the SOAP mechanism of Couvidat (2015), I suggest moving the text describing the SOAP mechanism to after Wang et al. (2024) is referenced. Please then also define the SOAP acronym, while also highlighting that this mechanism concerns the gas-particle partitioning of only the organics. In addition, please also include reference to the thermodynamics module that is used for the gas-particle partitioning of the inorganic species (which I assume is ISORROPIA).

Additional information is now provided in the revised version: "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."

2) As a general note, please do not forget to update the Wang et al. (2024) reference to include a link to the now published DOI.

Done in the revised version.

3) Please specify the CHIMERE model version used for this study.

The CHIMERE model version used in this study is that of Wang et al. (2024) based on a coupling between CHIMERE (Menut et al., 2021) and the SSH-aerosol v1.3 aerosol model (Sartelet et al., 2020):

"..., 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)."

4) I would suggest moving the sentence "Boundary conditions were taken from CAMS CIFS global model simulations (Flentje et al., 2021)." to after the IFS acronym is defined, and to then define CIFS to highlight that these reflect boundary conditions for the chemical species.

The revised version now reads: "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)."

Section 3.3

5) I think this section, and the CHIMERE model comparison as a whole, could be restructured. While the introduction contains background information on why PM measurements are valuable assets in the validation of atmospheric chemistry-transport (CTM) models, the exact motivation of including a model comparison in the context of the CARA dataset could be more clearly defined. In its current form, it is unclear to me why the model configuration is described in section 2.5, as it does not seem to fit the narrative of the paper at this point.

I would recommend adding a Section 4 divided into subsections that 1) motivate why a model-to-measurement comparison adds valuable information/results within the context of the CARA dataset, 2) model description moved from section 2.5 to here, 3) discussion of the model-to-measurement results, targeting clear implications or recommendations for the model configuration/implementation (also following comments of reviewer 1), to support the conclusion regarding the usefulness of the CARA dataset. A clear motivation for this model comparison should then also be

included in the introduction of the paper (the CHIMERE model is routinely validated against (organic and inorganic) PM observations, also in France within the context of the referenced RI-Urbans project, so what makes the lessons that can be learned from the CARA comparison especially novel?). In the current manuscript, this motivation is mostly limited to the final paragraph of the conclusion.

We thank the reviewer for highlighting that the motivation of this section did not appear very clearly in the submitted version. We have expanded the discussion regarding the model-to-measurement results in new section 4, presenting motivations, model description and results, which now reads:

"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

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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."

We have also added an additional paragraph in the Introduction to better clarify our objective L99:

"... They are also vital for evaluating and validating regional air quality models through comparison exercises, examples of which are also discussed in this article using CHIMERE CTM model simulations. Indeed, the CHIMERE model is routinely validated against observations, and the online data from the CARA program play a crucial role in France for the continuous enhancement of CHIMERE, resulting in more accurate forecasts."

6) In the context of the current work, I don't see the added value of including the comparison to the four filter samples (outside of the CARA dataset) shown in Figure S11, and would suggest leaving this out altogether. If the authors nevertheless decide to include the filter sample measurements, I would suggest including a comparison and discussion of why and how these measurements are different than those made during the CARA program elsewhere in the text, and not in the model-to-measurement comparison section. Going by Fig. S11, the difference in these co-located measurements seem very large to me (as is also apparent from the reported difference in correlation statistics with respect to modeled concentrations), in my opinion warranting a detailed discussion.

The offline measurements from filter samples are also part of the CARA program (Favez et al. 2021). We have added this example of comparison to show the importance of online vs offline measurements in validating CTM models.

We have added this information in L574: "Furthermore, we could compare the model results with offline chemical information from filter samples collected in the submicron aerosol fraction at four sites in 2018 within the CARA program."

7) Figure 7. Why is the model-to-measurement comparison limited to summer and winter? Currently it reads as if this is a limitation of the model, but if a simulation for the year 2018 was performed, surely this should not be a problem? The spring and autumn seasons could at least be included in the supplementary material for consistency with the seasons shown in Figure 5.

We have now added the other two seasons in the SI, Figure S11.
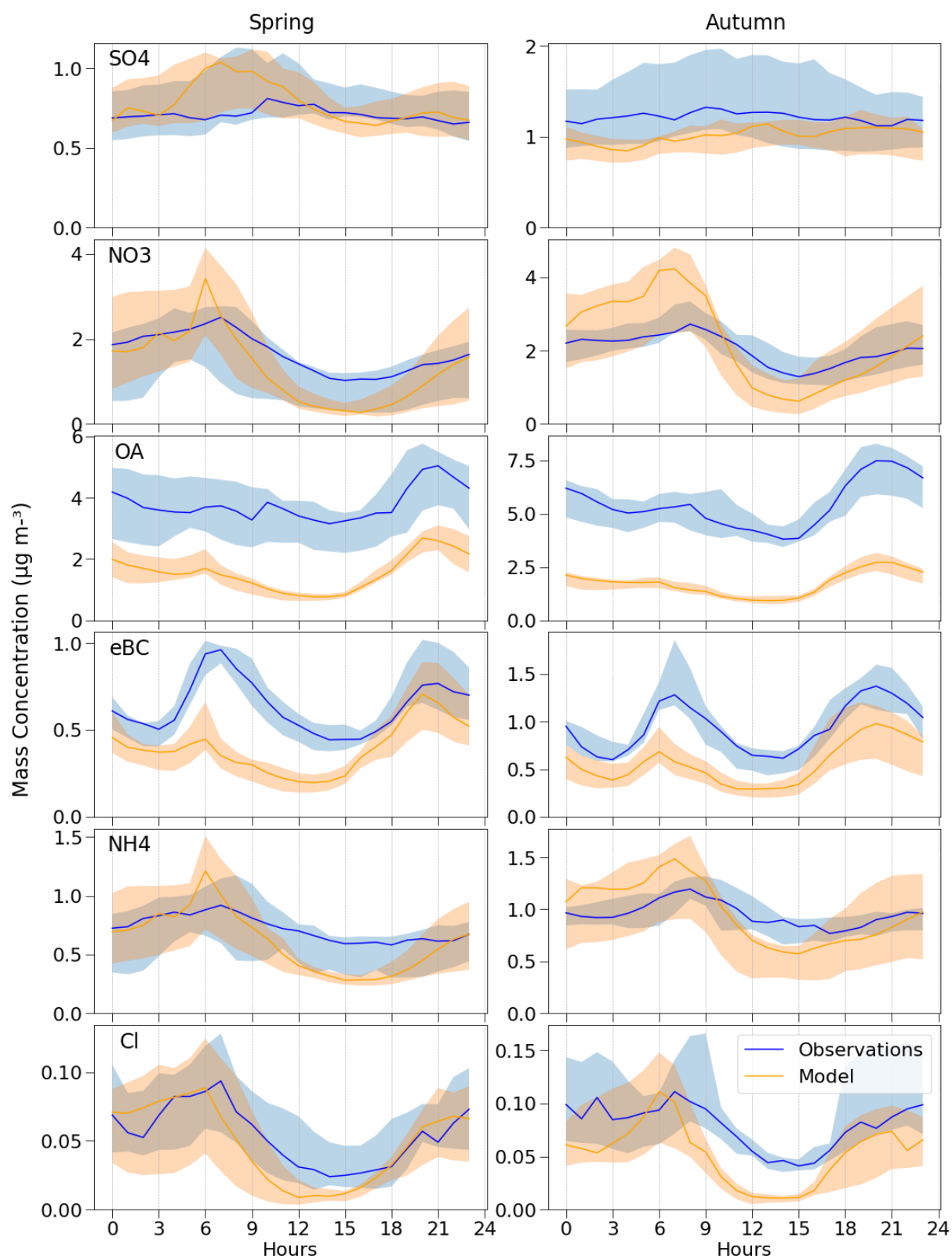
Data (availability)

8) A quick scan of the data reveals that there are many data entries for Cl and NH4 having values of 0.0055 and 0.142 ug/m3, respectively. Since this corresponds to exactly half their detection limits (defined as 0.011 and 0.284 uh/m3 in the readme file, respectively), I am not entirely sure what the origin of these numbers could be. Are they result of reaching the detection limit together with some kind of upper limit on the CE? For the CE the lower limit is specified at 0.5, but with the recurring entries scaling as 1/2 (=1/CE) times the detection limit, it would seem like there is also an upper limit of CE = 2 in place? But surely the CE can't be greater than 1?

Indeed, as mentioned in the main text, if the concentrations were between -3×DL and DL, they were replaced by the values of DL/2. (EPA QA/G-9, 2000, Hjellbrekke, 2001).

9) As a general comment, I would recommend explicitly specifying the units (I assume ug/m3) in both the measurement dataset as well as the readme file.

Yes, exactly, we have added this information in the readme file.

10) If the ATOLL (Lille), Marseille, and Paris data described in the manuscript are derived from the same ACSM measurements as those described in Chen et al. (2022) (their Fig. 2), I think this should be mentioned somewhere in the manuscript text.

In Chen et al. (2022), only one year of ACSM measurements (2016-2017) for these sites (ATOLL, SIRTA, and Marseille) was included, and the article focuses instead on the organic fraction and its sources. We have added this in the text, section 2.1, L110: "A one-year (2016–2017) dataset of ACSM measurements for ATOLL (Lille), SIRTA (Paris), and Marseille Longchamp was previously integrated into a multi-site European study (Chen et al., 2022)."

11) Since the CHIMERE model calculations are discussed in the paper, I think the relevant model outputs should also be included in the zenodo upload, even though they are not part of the CARA dataset. (at least following the submission guidelines for Geophysical Model Development – I can't seem to find whether it's different for ESSD).

We have uploaded the CHIMERE model simulations as well in the Zenodo link: https://zenodo.org/records/13318298.

12) The current link to the zenodo page containing the dataset points towards Version V1 which has restricted access.

Version V1 was not working and was replaced with a new version that opens directly with the Zenodo link provided in the revised version: https://zenodo.org/records/13318298.

Other comments

13) I suggest removing "Such chemically-speciated multi-year datasets have significant value for the scientific community, offering opportunities for future research, including source apportionment studies, trend analyses, and epidemiological investigations. They are also vital for evaluating and validating regional air quality models. In this regard," from the abstract and instead leaving it for the introduction, where I think this type of information is more fitting.

We have moved it to the end of the introduction in the revised version: "The datasets presented here are made fully available for complementary research activities, including trend analyses and epidemiological investigations. They are also vital for evaluating and validating regional air quality models through comparison exercises, examples of which are also discussed in this article using CHIMERE CTM model simulations. Indeed, the CHIMERE model is routinely validated against observations, and the online data from the CARA program play a crucial role in France for the continuous enhancement of CHIMERE, resulting in more accurate forecasts."

14) Since the CARA measurements are presented as PM1, I would suggest also stating this in the title (rephrasing from "fine PM").

The CARA program includes not only PM_1 measurements but also eBC measurements and filter analyses, which are both sampling the $PM_{2.5}$ fraction, which is why we have used the term "fine PM".

15) A brief explanation of how the ACSM measurements of PM1 relate to total PM2.5 might be good in the context of air pollution (typically measured in PM25) early on in the text, for example by referring to the discussion on lines 270-280.

We have added these lines in section 2.4: "PM_1 is a significant fraction of $PM_{2.5}$ especially in Europe (Putaud et al., 2004), understanding the composition and concentration of PM_1 is therefore essential for assessing the health risks and wider environmental impacts associated with $PM_{2.5}$ exposure".

16) Line 368. I think the difference in eBC between Chen et al. (2022) and the results of the current work is an important result. While some of the reasons behind this are discussed in section 2.3.2, I think it would be good to expand upon this (e.g., can it be verified which method is more correct? What is the relative importance of the MAC vs. harmonization factor?). Not in the least because Chen et al. (2022) is also a recent dataset, with many of its measurements and instrumentation participating in the same RI-Urbans project referred to in section 2.3.2, but also because the reported (methodological) 41% reduction in eBC levels strike me as very large.

Please refer to comment #10 from Reviewer 1:

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. The use of both parameters (H and MAC_ACTRIS) to calculate eBC reduces eBC levels in comparison with the instrument outputs (Zanatta et al., 2016).

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)."

17) L456. The increase in OA/eBC ratios during the day in summer seem to be the result of decreasing eBC concentrations rather than increased summertime biogenic (S)OA concentrations, as argued for in the text. For example, in Figure 5 bottom-left, the summertime OA diurnal profile remains almost perfectly flat, while that of eBC shows a noon-time minimum.

During summertime, the elevation of the boundary layer height would decrease the mass concentration in the afternoon of all pollutants depicted in µg m^{-3}, including eBC, and thus a "flat" variability of OA is indicative of net daytime enhancement. Thus, the increase in OA/eBC ratios could be explained by biogenic SOA formation in summer.

18) Line 123. Please define the acronym 'NR' (in NR-PM1) here.

Done in the revised version: "… non-refractory submicron aerosols NR-PM$_1$ …"

19) Line 149. I would suggest adding "(as discussed below)" behind "Middlebrook et al. (2011)" to highlight that the Middlebrook algorithm is discussed in more detail further on.

Added in the revised version.

20) Line 252. I would suggest leaving out the words "carefully" and "thoroughly" to make the wording of this sentence more scientific.

Done in the revised version.

21) L533-541. In the second paragraph of the conclusion, it is noted that OA is the predominant compound for the highest concentration levels in summertime at all sites, probably due to photochemical production. It is not clear to me where exactly this conclusion can be drawn from. In Figure 4, OA shows a pronounced summertime minimum, consistent with the daily mean (and nearly flat diurnal cycle) of OA being the lowest in spring and summer as shown in Figure 5. From Figure 4 and 5, it seems instead like the other species are low during summer, rather than OA being high.

Indeed, we have drawn this conclusion from Figure S4 in the SI where we present the relative contribution of chemical species as a function of PM$_1$ percentiles by season. We have added this in the conclusion: "Across all sites, OA is the predominant compound, with a mean concentration of 4.7 µg m^{-3} (43-60 %) in PM$_1$, followed by NO$_3$ (15-30 %), SO$_4$ (8-14 %), NH$_4$ (7-13 %), and eBC (5-11 %). Stations in central and southern France exhibit higher OA mass concentrations (5.3 µg m^{-3}), likely attributed to more pronounced photochemical formation processes. Such secondary processes may also explain that OA is the predominant compound for the highest concentration levels in summertime at all sites (Figure S4)."

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