

Correspondence to Anonymous Referee #2

The authors express their gratitude for the valuable feedback provided by Anonymous Referee #2. This document includes responses to each comment, with the referee's comments presented in blue font and the authors' responses in black.

Biomass burning emission is a hot topic in Air Quality in Europe. Biomass burning, mainly related to residential heating has recently increase due to the incentives to reduce greenhouse emissions. These emissions can be very important in medium size cities and in rural areas, and may have impact on both health and climate. As shown in the present article, this can be of great interest in rural areas frequently affected by thermal inversions. Moreover, there is growing interest in evaluating the optical properties of carbonaceous aerosols emitted by biomass combustion. The manuscript corroborated the importance of this source at rural areas and demonstrates the influence of coating of BC by OA in absorption and therefore on atmospheric warming.

This is a 2 months period campaign carried out in the village of Retje, in Slovenia. A complete set of instrumentation was settled at the village and at a reference location, 150 m higher. Instruments comprised: Aethalometers, MPSS, and CPC. Ions and EC/OC were determined at filters collected by high volume samplers. At the village site a total carbon analyzer was also used.

The paper is of interest and deserves to be published in ACP although there are some aspects that can be improved, mainly related to the uncertainty in the estimation of OA.

As stated in the manuscript, estimating OA hourly concentrations by subtracting BC and ions (measured in $PM_{2.5}$ and PM_{10} , respectively) from the PM_1 mass calculated from MPSS could be the largest source of uncertainty: 1) by the different sizes measured / sampled; 2) the MPSS in Retje measured from 10-800 nm; 3) because the ions were offline estimated in PM_{10} filters collected every 12h and a constant contribution of ions to PM_1 has been assumed, affecting the time variation of OA. Ions and EC/OC mainly concentrates in PM_1 but presence in the coarser fraction cannot be discarded. It is true that there is a very good correlation between PM_{10} and PM_1 derived from MPSS, indicating 90% of PM_{10} is in the PM_1 fraction as an average; but in some cases, with high PM concentrations, PM_1 accounts for around 70% of PM_{10} and then there is an important contribution of coarse PM that will affect the OA estimation. The authors compared OA_{MPSS} and OA_{TCA} and concluded that the good correlation corroborates the adequacy of the method used. However, it must be considered that, in both cases, OA/OC ratios used have been estimated by comparing the OA estimated from MPSS with the OC of filters. Therefore, the good correlation between OA_{MPSS} and OA_{TCA} only demonstrates a good correlation between OC_{tca} and OC filters, but does not provide evidence on the suitability of the method used for estimating OA.

This uncertainty in the estimation of OA may have a high impact on the results and conclusions. Thus, it will influence the estimation of MAC_{OA} . Then, I considered that more info about OA uncertainty should be provided.

Response: Thanks for your important observation regarding the OA mass concentrations. We recognize that the methodology employed in the OA calculation implies specific assumptions that are sources of uncertainty in the aerosol properties reported, like MAC_{OA} . To address this point, we have incorporated uncertainty calculations in the manuscript using error propagation. For the OA uncertainty, the individual sources of error contain the calculation of PM_1 mass concentrations from the PNSD, which include the MPSS deviations and aerosol density, the eBC uncertainty, and the contribution from the determination of inorganic aerosol mass concentrations in PM_1 (scaling fraction from PM_{10} to PM_1 , and uncertainties from the analytical method).

The PM_{10} uncertainty is taken as 17 % based on the calculations of Buonanno et al. (2009) and the MPSS intercomparison in the laboratory. This estimation accounts for contributions from the sampling flow rate, the volumetric diameter, the diffusion efficiency corrections, and the particle density. The uncertainty of the eBC mass concentration is assumed to be 5 %, corresponding to the EC uncertainty, as eBC was normalized to EC. The uncertainty of EC corresponds to the reproducibility of the thermo-optical analysis following the EUSAAR protocol. The uncertainty contribution of the insoluble inorganics was assumed to be 20 %, accounting for the PM_{10} - PM_{10} scaling and repeatability, an important contributor to uncertainty in inorganic content determination, as shown in Leiva et al. (2012). The final uncertainty for the OA mass concentration was 22%. Additional details are given in the new section 3.3.

Regarding the comparison between OA_{MPSS} and OA_{TCA} , we agree that the use of a local OA/OC ratio results, in the end, in a biased comparison. Consequently, we have opted to show a comparison between OA_{MPSS} and OC from TCA (OC_{TCA}) and filters ($OC_{filters}$) (see updated Fig. S1). The agreements in both comparisons and the resulting OA/OC ratios (slopes) show that the estimated OA_{MPSS} follows a realistic trend of the organic aerosols in the study site and suggest that the hourly OA_{MPSS} are comparable to independent methods. The differences in the estimated OA/OC ratios are related to the distinct sampling periods (1-hour averaged data from TCA and 12-hour data from filters) and techniques (Brown et al., 2013). However, both OA/OC ratios fall within the range of reported results in previous studies (Srinivas and Sarin, 2014; Xing et al., 2013). These changes were included in section 2.3 (see lines 238 to 244) and Fig. S1 in the Supplement.

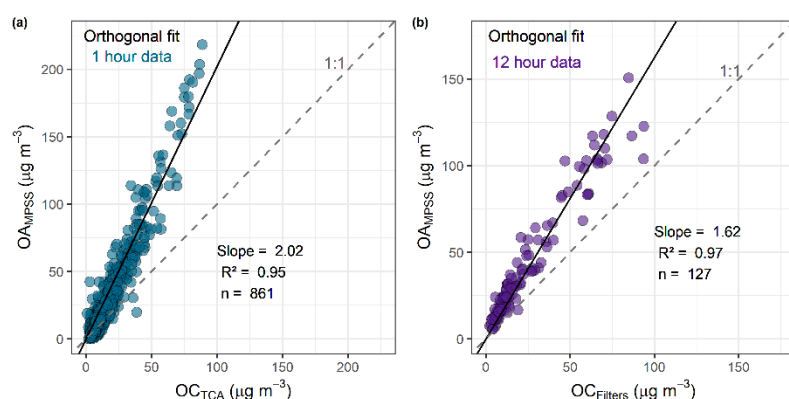


Figure S1. Scatter plots and orthogonal regressions (solid black lines) for the comparison of (a) OA_{MPSS} and OC_{TCA} , and (b) OA_{MPSS} and $OC_{filters}$.

Minor corrections

- Line 139. Add reference for TCA

Response: The reference to the TCA was included in the text (see line 148). Additional information about the instrument's principles of operation was also included in section 2.2.

- Line 180. This Table can go to Supplementary. Information on inlets size cut should be added

Response: We appreciate this suggestion; however, we respectfully prefer to keep Table 1 in the main text as a summary of the instrumentation used during the campaign. Nevertheless, information about the inlets is included in section 2.2 (see lines 155 to 158) and is also mentioned in Table 1 now.

- Line 270: Table 2. Can you add the % of hours for each category during the sampling period? Or just shortly describing the frequency of the stability categories in the text.

Response: The percentages of occurrence for each condition of atmospheric stability are included in Tables 3 (hourly basis) and 4 (predominant during periods of 12 hours). A mention on the frequencies of each category is given in lines 334 to 335. For hourly measurements, the percentages of occurrence were 28 % for strong inversion, 31 % for weak inversion, 5 % for neutral atmosphere, and 36 % for unstable atmosphere.

- Line 282 (and Fig.3): Does OA refers to OAmpps? It should be clearly stated that OA refers to OAmpps in the manuscript.

Response: Thanks for this observation. In effect, the term OA refers to OA_{MPSS} . We have indicated that OA_{MPSS} is called OA in the manuscript (see line 245).

- Lines 287-289: PNC is very similar for strong inversion and unstable atmosphere.

Response: In this paragraph, we compared the maximum particle number for both categories of atmospheric stability. However, the mean concentrations are significantly different: for strong inversion, the mean N was 17×10^3 particles cm^{-3} , while for unstable atmosphere, the mean N was 6.1×10^3 particles cm^{-3} . We have modified the text and reported mean values of N in section 3.1 (see lines 317 to 318).

- Line 312-317. Little discussion about ΔPNC and $PNSD$; I understand this is not the topic of the articles. PNC measurements have been mainly used for deriving $PM1$ and hourly OA. However, I would add an explanation about similarity of ΔPNC for N_{10-50} during the three categories

Response: We agree with this comment and have included additional text commenting on this matter (see lines 346 to 350). In summary, we observed median ΔN_{10-50} of 12×10^2 , 9.8×10^2 , and 9.6×10^2 particles cm^{-3} for strong inversion, weak inversion, and unstable atmosphere, respectively. From strong to weak inversion ΔN_{10-50} decreased by 18%, while this delta remained practically constant between weak inversion and unstable atmosphere. In general, ΔN_{10-50} exhibited the lower change among the stability conditions, given that from strong inversion to weak inversion, ΔN_{50-100} reduced by 50 % and $\Delta N_{100-600}$ by 60 %.

We hypothesize that the almost constant ΔN_{10-50} is explained by the predominant sources of ultrafine particles: secondary aerosol particles, sea salt, and traffic emissions (Leoni et al., 2018). All these three sources might have an impact on the local and background concentrations at the study site. From the hourly profiles of N_{10-50} , peaks can be observed during typical commuting hours (6:00-09:00 and 16:00-17:00) at Retje and the background stations (Fig. 1).

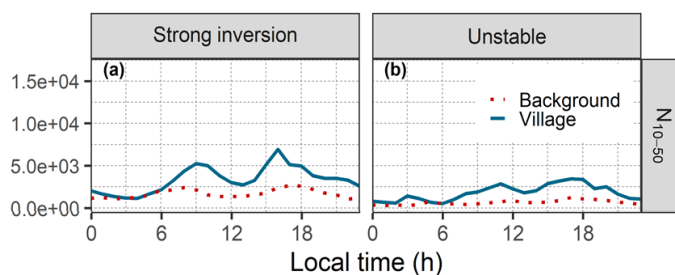


Figure 1: Hourly variation of the N_{10-50} in the village and the background stations.

- Figures 4 and 5. Captions: Please, remove “black dots” at the end of the caption. Check whiskers: do represent 25-75%?

Response: Thanks for this remark. We have corrected the caption in Fig. 4 by removing “black dots.” The whiskers represent the minimum and maximum values without outliers; the distances between the

whiskers contain 25 % of the data at each side of the distribution. The captions in Figures 4 and 5 were updated to avoid misunderstandings.

Note: In Fig. 4, the changes in OA (Δ OA, Fig. 4a) were excluded since, only in this section, the mass of PM_{10} at the background station was calculated and used to estimate OA. However, the maximum mobile diameter covered by the MPSS in the background (600 nm) does not cover PM_{10} in its entirety, making it inaccurate to calculate PM_{10} and, therefore, OA mass.

- Figure 6. I do understand the increase of absolute concentrations during strong inversions. How do you explain the increase of the relative contribution of BrC with respect to BC_{total} ? Is it because strong inversions are mainly produced at night when domestic heating emissions are more important? Or because you assume all BB emissions are local while traffic emissions are also external? Based on the results obtained in the paper, do you believe this source apportionment is realistic? Have you compared with BC SA at the reference site?

Response: We believe the larger fraction of absorption attributed to BrC during strong inversion is a combined result of local emissions and accumulation because of the weather conditions, i.e., during strong inversion, there might be an increase in local wood burning due to lower temperatures, but the accumulation of the local pollution is higher due to reduced vertical mixing and ventilation. The last minimizes the influence of external sources of pollution, and so we hypothesize that the vast fraction of BrC proceeds from wood burning at the study site. On the other hand, during unstable atmosphere, there is a higher chance that local pollution gets diluted and also mixed with external sources such as traffic from national and regional routes or long-range transported aerosols. However, there are traffic emissions at the study site from local households, which might eventually contribute to local measurements, though this contribution is rather small (less than 100 vehicles circulating daily) and concentrated during specific periods of the day (Glojek et al., 2020, 2022).

We do have confidence that the absorption apportionment is representative of the dynamics occurring in the study site. Using a different approach for the same study site in winter, Glojek et al. (2020) found an average contribution of 63 % from biomass burning to eBC mass concentrations, compared to 37 % for traffic emissions.

Respect the last question on this comment, we are unsure about the meaning of SA in the referee's question. We assume this might be scattering albedo and, consequently, we have estimated the SSA from our Mie modeling results. The values of SSA were significantly lower than the usual values reported in the literature. At 470 nm, we found a mean SSA of 0.48 for strong inversion and 0.51 for unstable atmosphere. At 660 nm, SSA were 0.61 and 0.63 for strong inversion and unstable atmosphere. The values of SSA might not be reliable enough to draw significant conclusions. In general, from other studies we know that particles with a comparatively higher fraction of OC (and BrC) than EC, have SSA closer to 1 at 660 nm and close to 0.9 at ~ 400 nm (Pokhrel et al., 2016).

References

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