

This paper presents an analysis of the impact of ground temperature inversions on total particle mass concentrations and in particular on black carbon. The experiments were carried out in rural Slovenia, with emphasis was on wood combustion aerosol pollution. Overall, the paper is well written and the experimental approach is well described. I would recommend its publication in ACP after some revisions:

**Comments:**

- How is the profile along the fixed route? Why is only the profile perpendicular to the route shown?

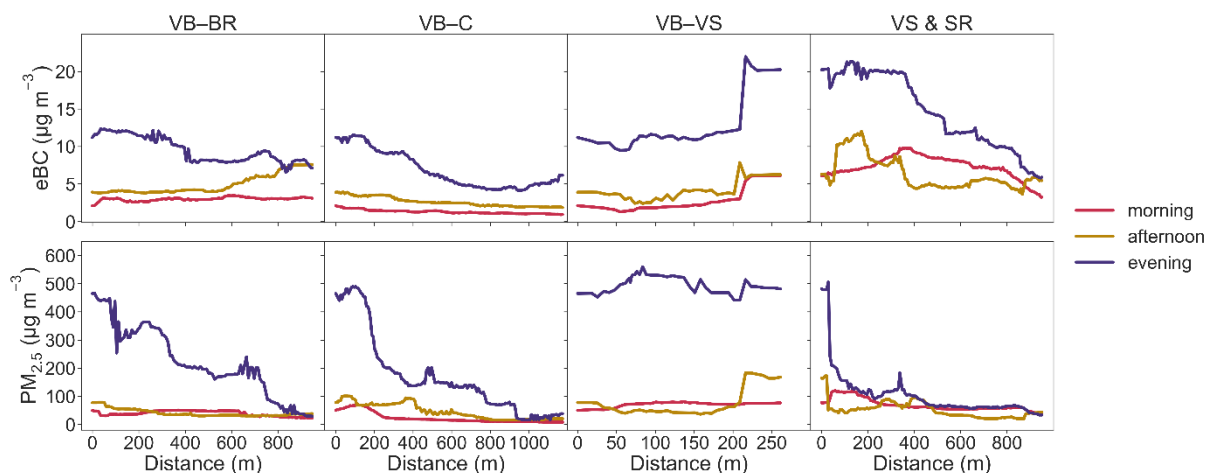
**Response:**

Thank you for the question, we have included the horizontal gradients in the manuscript, under the Table 4 of 3.2 Chapter.

**Changes in the manuscript:**

At the end of the chapter 3.2:

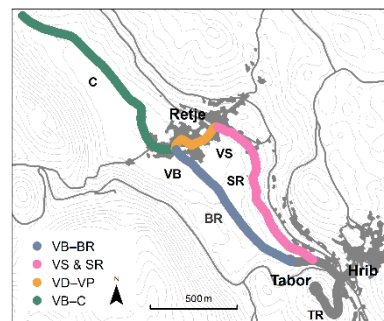
A deeper analysis of the pollutants' spatial distributions during temperature inversions is shown in Figure 4. The determined line segments along the fixed route inside the Retje hollow are listed and described in Table 4 with their spatial locations marked in Figure 5. Here, the goal was to show horizontal variability of eBC and PM<sub>2.5</sub> along the hollow with respect to different times of the day. To this end, line segments at similar relative heights were selected (a maximum difference of 30 m per route segment; see Table 4).



**Figure 4: Variability of eBC (top panels) and PM<sub>2.5</sub> (bottom panels) mass concentrations during morning (6:30–8:30 LT, red line), afternoon (12:00–14:00 LT, yellow line) and evening (17:00–19:00 LT, purple line) winter-time temperature inversion along different parts of the fixed route in the Retje hollow.**

**Table 5: List and description of the different route segments along the Retje hollow.**

| Location code | Descriptions                                                  | Altitudes (m) | eBC/PM <sub>2.5</sub> |
|---------------|---------------------------------------------------------------|---------------|-----------------------|
| VB-BR         | Bottom route in a village Retje-Hrib direction.               | 705.9–741.5   | 0.12                  |
| VB-C          | Line segment in a village Retje-chapel st. Florjan direction. | 706.7–728.9   | 0.11                  |
| VB-VS         | Bottom part of village Retje in a west-north direction.       | 707.1–719.2   | 0.07                  |
| VS & SR       | Slope route in a village Retje-Hrib direction.                | 720.3–758.4   | 0.16                  |



**Figure 5: The location codes assigned to the line segments of the fixed route.**

eBC and PM<sub>2.5</sub> mass concentration variability trend differs throughout the route and it is day time dependent as well. Namely, mass concentrations of both pollutants are 2- to 4-times higher during evening temperature inversions as compared to inversions in the morning and in the afternoon. This applies to all locations inside the hollow. Yet, the

difference between the run times is the highest in Retje village, with up to 5- and 7-times higher evening eBC and PM<sub>2.5</sub> levels, respectively.

Unlike the PM<sub>2.5</sub>, the eBC trend along the line segment VB–BR depends on the time of the day. In the morning, from 6:30 to 9:00, eBC levels with the mean value of 3 µg m<sup>-3</sup> remain constant along the whole bottom route (BR), whereas around noon and in the early evening, eBC mass concentrations vary. As seen with the PM<sub>2.5</sub> levels, early evening eBC mass concentrations increase from 6.6 µg m<sup>-3</sup> to 11.6 µg m<sup>-3</sup> towards the village of Retje. In the other direction, from the village of Retje to st. Florjan chapel (VB–C), levels of both pollutants decrease, with the highest drop observed during evening temperature inversions, particularly in PM<sub>2.5</sub>.

Horizontal gradients of the pollutants' mass concentrations differ the most at Retje village located at the bottom of the hollow (VB–VS) and at the slope street in the direction from Retje village towards the settlement Hrib (VS & SR). This relates to morning and evening temperature inversion runs, while for the afternoon runs variability of the pollutants is similar. In the morning and in the evening, PM<sub>2.5</sub> mass concentrations over the whole village at the bottom of the Retje hollow are elevated and steeply decline further away from the village, especially in the evening. In the afternoon, however, greater mass concentrations were measured along the village on the slope (VS). This is seen in Figure 4, line segment VB–VS, as a sharp increase in PM<sub>2.5</sub> mass concentrations at a distance of 200 m away from the route segment starting point. The decrease of eBC along the slope road (VS & PC) from Retje village towards Hrib is smaller than that of PM<sub>2.5</sub>. Moreover, during morning runs, eBC mass concentrations in the middle of the road segment increase, i. e. from 4.4 to 9.2 µg m<sup>-3</sup>.

Line 596–598: Moreover, the high correlation between eBC and PM<sub>2.5</sub> mass concentrations along the whole hollow indicates homogeneous atmospheric conditions with the prevalence of one emission source. However, different morning and evening horizontal gradients of eBC and PM<sub>2.5</sub> were observed during temperature inversions in the village of Retje. We partially interpret this as secondary aerosol formation (SOA).

There are two Figures S12.

**Response:**

Thank you for this comment. We do see oversight and we have changed wrong Figure S12 into S11 in line with the pdf\* document.

- References to Sections within the text should be checked (e.g., line 315 and 435)

**Response:**

Thank you for the noticed incorrect citations. We have checked them and corrected accordingly.

**Changes in the manuscript:**

Line 315: »Concerning the second goal, mobile measurements were spatially averaged according to the method described in Alas et. al., 2019a and Alas et al., 2019b.«

- How strongly are the eBC vertical profile influenced by direct eBC emissions? Is an error estimation possible?

The difference between group means is indeed statistically significant, by about 0.3 of standard deviations lower eBC-MH compared to  $\theta$ -MH. Population value of a slope between the  $\theta$ -MH and eBC-MH, is likely to fall between 0.52 and 0.92 (95 % confidence interval). The relatively wide interval suggests a high degree of uncertainty of our method. Yet, we cannot conclude that an effect is important because the p-value, from which we determine significance, is affected by a small sample size (22 runs with both mobile temperature and eBC measurements). From frequency distribution of the  $\theta$ -MH and eBC-MH data shown in the subplots of upgraded Fig. S13 in the Supplement (blue histogram for the  $\theta$ -MH and gray for the eBC-MH) it is evident that there is a peak in the number of runs with  $\theta$ -MH of 60 m, while for the eBC-MH, most of the runs have MH of 40 and 70 m. The former eBC-MH represents predominantly early evening runs and the latter morning runs.

Despite the method shortcomings, the difference between the  $\theta$ -MH and aerosol-MH over complex terrain was observed in some other studies as well (e. g. De Wekker et al., 2004; Ferrero et al., 2010; Lang, Gohm, Wagner, 2015). Furthermore, Seibert et al. (2000) states that there is a lower agreement between the two methods for very low MH. Comparison with values of correlation coefficient and regression slope reported by e. g. Ferrero et al.

(2010, 2011) show that the degree of correlation for our study is lower. However, the correlation coefficient agrees well with the one reported for the profile with timing and resolution comparability issues (Ferrero et al., 2011). Besides, root mean squared error (RMSE) indicates that our model misses actual MH values by less than 12 m (RMSE=11.43 m), which is approximately from more than 10 to 65 m less than reported in Ferrero et al. (2010, 2011). Mean absolute percentage error (MAPE) of the linear model's predictions are, on average 12.3% off from actual value. Moreover, there are more negative errors than positive, indicating a systematical underestimation of the eBC-MH. Considering absolute and relative deviations of the MHs for every run separately, the highest discrepancy was observed for the 171229 A and for 171216 C. The latter being listed under the Fig. S13 in the Supplement as a special case.

One of the major limitation of our model is that the dataset contains only a small number of different values, since the MHs were determined only during temperature inversions at the specific times in a day. In this case, also using for example, the bootstrap sampling, does not improve the sampling distributions of the variables and thus the linear regression model. As we explain below in more detail, local spatiotemporal factors influencing the eBC profiles are interconnected and thus, their separate contribution in the frame of this study is not possible. Besides, for safe interpretation, we lack of additional data (e. g. traffic counts, one of established vertical profile methods, wind measurements and/or pseudo-vertical measurements along the other parts of the hollow).

The estimated uncertainty of the method used in our study is mainly result of:

1. Different timing and resolution of the  $\theta$  and eBC measurements.
2. Lower comparability of the two methods during weak inversions and during mixing heights with stratified PBL structure.
3. Influence of the route characteristics (land use and cover, position in the hollow, terrain configuration and thus local flows) combined with activity of the emission sources (RWC, morning traffic).

As we state in lines 306–308 in the manuscript, resolution of temperature measurements was 2 min, while resolution of aerosol measurements was 10 s. This leads to the reduced accuracy of  $\theta$ -MH. Namely, in 2 min time approximately 12.5 m distance and minimum 2 and maximum 20 m relative height had been walked from Hrib up to Tabor hill (13.5 m on average). This indicates that the  $\theta$ -MH error could be as high as 20 m. Nonetheless, this is still in line with a recommended resolution range for MHs lower than 250 m (Seibert et al., 2000). This ensures that relative uncertainties do not exceed 20 %. Here, it should also be mentioned that temperature sensor time was not synchronised with the time of aerosol measurements, which might, to a lesser extent, attribute to time difference as well.

Secondly, we found that the relative difference between the two methods increases up to 54 % in case of weak temperature inversion. While during pronounced temperature inversions, the relative difference is much lower, around 15 %. This is due to less significant differences in the vertical profile during temperature inversions with near-neutral  $\theta$  gradient, which makes identification of the MH more difficult. The comparability of the two methods differs between different run times as well. Overall, the agreement is better for the morning and afternoon temperature inversion runs than for the early evening runs. Higher discrepancy for the early evening runs could be the result of stratified nocturnal PBL with a stable surface boundary layer (SBL) formed at its bottom part. This is consistent with findings of studies described in Seibert et al. (2000), by Gregorič et al. (2020) and Ferrero et al. (2012).

Thirdly, an important and already mentioned contribution to the uncertainty of the method arises from the route specific measurements. Activity of emission sources along the fixed route, mainly residential wood burning (RWC) and also morning traffic, have an impact on the obtained vertical eBC profile and thus, on the determined MHs. In addition, the influence of land use and cover, the terrain configuration with related slope winds have a stronger impact as if we would use “standard” vertical measurement method. In particular, the most often occurring morning MH of 70 m, is very likely impacted by local factors. Namely, the height corresponds to the drainage area at this part of the hollow with the influence of locally induced flows. Moreover, above this height there are much less emission sources than in lower parts. These factors overlap and are interconnected, hence, the determination of various contributions is not possible.

A limitation of our method that we need to mention as well, is a time resolution of the obtained MH. Instead of recommended 1 h or less (Seibert et al., 2000), MHs in the study have a time resolution of 1.5 h (duration of the mobile measurements run). Hence, the evolution of the MH in the morning and in the early evening might not be adequately described.

With the following steps we tried to address some of the listed challenges arising from vertical mobile measurements along the slopes:

Firstly, single events, such as construction work, chimney plume or passing a heavy-duty vehicle were minimized by averaging 1-s raw data of AE51s to 10-s medians as suggested in e. g. Peters et al., 2013; Alas et al., 2018; 2019a. With the latter, the influence of AE51s measurement noise was reduced. As shown by Alas et al. (2021), however, it could still present an issue, particularly in areas with eBC levels smaller than  $4 \mu\text{g m}^{-3}$ . However, the vertical eBC profiles presented in the paper are an average of many measurements rounded to the nearest 5 m relative height. Hence, the effect of the noise on the reported mass concentrations is further reduced. The effect of data aggregation is demonstrated by the standard deviation of the vertical eBC mass concentrations in Fig. S13. It is evident that measurements in areas with a higher concentration level have a higher variability than in the lower levels. Moreover, the special single events, which could have biased the results, were noted in the mobile measurements log-book and taken into consideration.

Secondly, with a circular fixed route, the obtained vertical profile of the hollow is a result of data points of different parts along the hollow. There is an overlap between different line segments and thus, up to 60 m relative heights, the profile is not route part specific (see Fig. S11 for the route parts height ranges). However, from there to the top of the hill at 115 m height, there is only one line segment (part of H and TR). With crossing the latter at least twice per every run (up and down the hill and after completion of some runs once more) we averaged out the single events and obtained representative distribution of eBC concentrations for the south east side of the studied area. Yet, we cannot claim that the results are valid for the other unpopulated south side of the hollow. Even if we had data from the other, unpopulated south west side of the Tabor hill, comparison and thus, the estimate of direct effects of local emission sources would not be sufficient, due to different terrain characteristics.

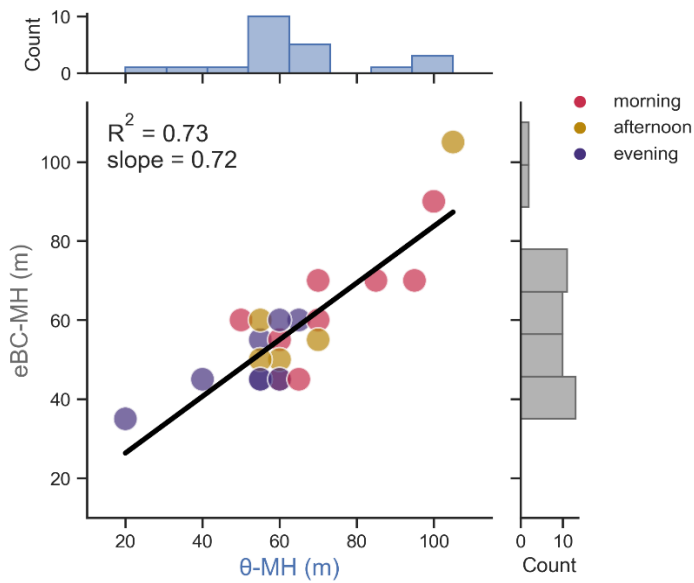
Lastly, for the MH determination, we have used data from the fixed stations as well. Besides  $\theta$ , we considered RH measured at the three stations (Retje village station, meteorological station Hrib and Tabor background station). A significant reduction in RH among those stations was used as a sign for the height of a mixing layer. Additional control parameter, whether the MH is below or above the Tabor hill, was a comparison between eBC mass concentrations at the Retje village station and at the rural background station on Tabor hill. As shown also in the study of Gregorič et al. (2020), higher concentrations in terrain depression than on top of hill indicate MH below the altitude of the hill site.

Alas, H. D., Stöcker, A., Umlauf, N., Senaweera, O., Pfeifer, S., Greven, S., Wiedensohler, A.: Pedestrian exposure to black carbon and PM<sub>2.5</sub> emissions in urban hot spots: new findings using mobile measurement techniques and flexible Bayesian regression models, *Journal of Exposure Science & Environmental Epidemiology*, December 2020, 1–11. doi:10.1038/s41370-021-00379-5, 2021.

Gregorič, A., Drinovec, L., Ježek, I., Vaupotič, J., Lenarčič, M., Grauf, D., Wang, L., Mole, M., Stanič, S., Močnik, G.: The determination of highly time resolved and source separated black carbon emission rates using radon as a tracer of atmospheric dynamics, *Atmospheric Chemistry and Physics*, 20, 14139–14162, doi: <https://doi.org/10.5194/acp-20-14139-2020>, 2020.

### **Changes in the Supplement:**

Upgraded Fig. S12. Frequency distribution of the MHs and identification of time of day (color-coded points) added to the Figure.



**Figure S12: Correlation between  $\theta$ -MH and eBC-MH for temperature inversion runs in December.**

- BC source apportionment reported in the cited manuscripts still show significant traffic contribution during certain periods during the rush hour period. Would be interesting to have some numbers also in the present manuscript together with a comment, e.g., line 585 what is the contribution of local morning traffic to total eBC?

**Changes in the manuscript:**

Lines 585–588: »Local morning traffic (people driving to a local school and to their workplace) contribute slightly as well, especially when it comes to eBC mass concentration levels (Glojek et al., 2020). During the peak hour, at 9:00, traffic accounts for 34 % of eBC in Retje (Glojek et al., 2020).«