

We thank for the comments of the Referee #2. The comments are numbered and each of them is followed by a response from the authors.

General remarks:

1) COMMENT: At several places in the text the authors refer to “good” and “bad” correlations. Please use the terms statistically significant or not accompanied by statistical tools (t-test).

REPLY: Referee is correct that terms “good” and “bad” were used vaguely in text. That has been changed. In most of the cases verb “correlate” was replaced by “agree” or “compare” that do not refer to statistical analysis. However, it should be noted that this manuscript reports a proper statistical analysis in Figs. 4 and 9, and in supplements in Tables 1 and 2 and Figs. S5 and S10.

2) COMMENT: In several figures especially in figs 1, 5, 6 and 7 please mark with a horizontal line the periods (I, II, III etc) referred in the manuscript, page 8283. It will greatly help the readers to follow the species variability.

REPLY: We used vertical lines in Figs. 1, 5, 6 and 7 to mark the periodization of the campaign.

3) COMMENT: Did the authors measured BDL variation? Figure 10 and Table 1 comment on the behavior of several compounds before and after its nocturnal break-up but no explanation on how this break was defined is given in the manuscript. See also paragraph 3.4 lines 17-21.

REPLY: Boundary layer mixing height was measured by a LD-40 lidar-ceilometer. An average profile for the development of mixing layer height was added to Fig. 10a. Additionally, a description of the lidar-ceilometer measurements was added to Section 2.5 (Experimental methods) and a sentence: “These days were selected based on the time series of the AMS species and the mixing layer height from the lidar measurements.” was added to Section 3.4.

Other remarks:

4) COMMENT: *Line 21, page 8272: The authors should better describe the aim of their work, the way they did is quite vague.*

REPLY: The aim of the work has been sharpened. See also comment #1 for Referee #1.

5) COMMENT: *Lines 8 and 22 pages 8274: Did the authors correct their OC and WSOC values for blanks or they consider them quite low.*

REPLY: OC, WSOC, inorganic ions and organic acids were all corrected for blanks by subtracting the concentration of back-up filter from that of the front filter. Sentences “OC and WSOC concentrations were corrected for blanks by subtracting the concentration on the back-up filter from that on the front filter.” and “Similar to OC and WSOC, inorganic ion and organic acid concentrations were corrected for blanks.” were added to text.

6) COMMENT: *Please specify? CE: As collection efficiency is species dependant why they don't correct values based on the relations with filters? My remark goes especially to sulfate showing the biggest discrepancy with filters (factor of 2). I don't see the interest in presenting sulfate levels of the order of 0,9 ug/m³ or lower which are definitively wrong for a continental European area. Modelers can make use of these results (line 6 page 8284) or simply by referring figure 2.*

REPLY: Referee is correct that the collection efficiency of the AMS depends on the chemical composition of aerosol. CE less than unity is mostly due to the bounce of particles from the vaporizer, which is a function of the aerosol phase and therefore also the composition of particles. Middlebrook et al. (2012) proposed a parameterization for CE that depends on the acidity and ammonium nitrate fraction of aerosol. That equation calculates the CE based on the chemical composition of aerosol but as all the measured chemical species are assumed be internally mixed the obtained CE is applied to all chemical species. What Referee is suggesting here is probably species dependent-CE. Unfortunately it wouldn't work here because there is no reason why sulfate would be in different particles from ammonium, nitrate and organics and therefore have different CE. The reason for the low sulfate concentrations from the AMS was probably technical issues. As already explained in Section 2.4.1 the vaporizer temperature was probably too low. Even though the sulfate concentrations and contributions are too low in Fig 2, we do not see a reason to remove sulfate from Fig. 2 as the

time series and diurnal trends are assumed to correct. Regarding the concern about modelers using the sulfate data from Fig. 2, we added a sentence:

“Note that sulfate concentrations from the HR-ToF-AMS were much smaller than those from the BLPI or PM₁ filter measurements probably due to the instrumental issues. See details in Section 2.4.1.” to Figure caption 2.

7) COMMENT: *Line 23, page 8283, Define stars and arrows at caption of figure 2.*

REPLY: A sentence “Asterisks and arrows in (a) present the cases with the nocturnal surface layer break-up discussed in Section 3.4.” was added to Figure caption 2.

8) COMMENT: *Figure 4, explain color scale with dates at figure caption.*

REPLY: Dates have been added to figure caption of Fig. 4.

9) COMMENT: *Line 27, page 8289, no measurements of solar radiation are available from a regional meteorological office? The word “supposed to be pretty steady” is not adequate.*

REPLY: Solar radiation measurements are actually available at the site and but there was no trend during the campaign that could be linked to the steady decrease of levoglucosan concentrations. A free-access website where quicklooks of these data are available: http://atmos.cr.chiba-u.ac.jp/html/gli_sfc/gli_bologna.html. Text “was supposed to be pretty steady” was replaced by “did not show any trend”.

10) COMMENT: *Line 14, page 8290, any explanation on the moderate correlation between biomass burning tracers and nss-K?*

REPLY: There can be several reasons for only the moderate correlation between biomass burning tracers and potassium. Potassium can have several other sources than biomass burning for example sea-salt or soil. Unfortunately we couldn't correct potassium for sea-salt contribution since we were not able to analyze sodium from the PM₁ filters due to high blank values. However, there shouldn't be much sea salt or soil in PM₁ size range. Also the biomass burning conditions can affect the ratio of levoglucosan to potassium. Frey et al. (2009) found that smoldering combustion increases the concentrations of levoglucosan compared to normal

burning conditions in a masonry heater whereas potassium concentrations decreased during smoldering.

11) COMMENT: *Lines 23-26, page 8291. Is there any correlation between HOA and NO_x? Please clarify as first sentence (line 23) says “strong correlation” and the second (line 25) “quite small”.*

REPLY: As shown in Table S2 in Supplements, the correlations (R) between HOA and NO_x, NO and NO₂ were 0.539, 0.542 and 0.463. Those correlation coefficient were quite small, but by comparing the time trends of HOA and NO_x/NO/NO₂ visually, we could identify several common features. In order to clarify the text, the sentence “Of all the PMF factors, HOA had the strongest correlations with gaseous NO_x, NO and NO₂.” was deleted.

12) COMMENT: *Line 7, page 8292. The statement “OOA-a is a clear regional component” is not clear why. The authors explain it further but here is not clear at all.*

REPLY: “OOA-a was a clear regional component” was deleted from the beginning of the chapter.

13) COMMENT: *Line 10, page 8297: N-OA change reported by authors is not clear in figure 10c.*

REPLY: We agree with the Referee that the change of N-OA is difficult to see in Fig. 10 c. Therefore the reference to Fig. 10c was deleted.

References

Frey, A.K., Tissari, J., Saarnio, K.M., Timonen, H.J., Tolonen-Kivimäki, O., Aurela, M.A., Saarikoski, S.K., Makkonen, U., Hytönen, K., Jokiniemi, J., Salonen, R.O. & Hillamo, R.E.J. 2009: Chemical composition and mass size distribution of fine particulate matter emitted by a small masonry heater. *Boreal Env. Res.* 14:255–271.

Middlebrook, A. M., Bahreini, R., Jimenez, J. L. & Canagaratna, M. R. 2012: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, *Aerosol Sci Technol.* 46:3, 258-271.