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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trace metal characterization of aerosol particles and cloud water during HCCT 2010" *by* K. W. Fomba et al.

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General comments: The paper is generally well written and the data presented in the paper are important for aerosol – cloud interaction studies. However, the parts related to general comments on aerosol origin and relations between individual species found by chemical analysis in aerosols have tendency to overstate what it is found in the data that are actually composed from few data points only. The correlations between different species in aerosol connected with anthropogenic emissions tend to be high as they are often driven by meteorological situation causing various atmospheric mixing. The changes in size distribution and concentrations between upwind and downwind sites are quite different from each other if one takes into account that these four events

were carefully selected from many sampling days.

- The interpretations based on the correlations have been reviewed and these sections have been revised accordingly. The discussion on the source apportionment based on the correlations has also been changed.

One would expect more detailed evaluation of connected meteorology or possible sources interacting with the transported aerosol at the downwind site to improve reasoning about these differences. Therefore, I think, the paper needs major revision before it will be suitable for publication in ACP

- A detailed evaluation of the connected meteorology and interconnection of the three sites have been presented by Tilgner et al. 2014. Therein, the general synoptic weather situations, including the mesoscale flow conditions, local meteorological conditions and local flow conditions, SF6 tracer experiments to access the connection of the air masses between the three measurement sites during this experiment as well as implementation of regional models to describe the general meteorological conditions during this experiment have been discussed.

The other minor comments are below:

The sampling dates and times of samples analyzed here should be given earlier than in the last picture description

- Sampling dates and times have been added at the beginning of the results section on Page 7 lines 19-21 and which now reads,.

"The presented results are focused on data obtained during four full cloud events (FCE); FCE1.1 (14/9/2010 11:00 to 15/9/2010 01:00), FCE 11.3 (02/10/10 14:30 to 19:30) , FCE 13.3 (10/6/2010 12:15 to 10/7/2010 3:15), and FCE 22.1 (10/19/2010 21:30 to 10/20/2010 3:30)."

p. 5, line1: the lowest cut point (stage 1) is missing in the list.

- The information about the lower cutoff has been added. The sentence now reads,

- "The collected particle diameter ranged from 0.05 to 10 μm with lower stage cutoffs (stages 1 to 5) at aerodynamic diameters of 0.05 μm, 0.14 μm, 0.42 μm, 1.2 μm and 3.5 μm, respectively."

p. 9, line 5-6: Entrainment of air above may also increase the concentration.

- We agree.. A corresponding sentence has been added and it now reads: "Entrainment of cleaner air from above might also reduce concentrations, or in the case of more polluted air mass from above, increases the concentration."

p. 9, line 27: There is apparent increase of Ni, Cr (at least 3 times what authors call Insignificant) and Cu concentration at GB site in FCE 11.3 sample (Fig. 1), but there is almost no comment about it.

- As suggested on P. 9 lines 8-11, local traffic emission may influence these concentrations. A corresponding sentence has been added to the manuscript which now reads.

Although entrainment of more polluted air mass from above may increase aerosol concentrations, the increase in the trace metal concentrations was not uniform for all investigated trace metals but rather more specific for these trace metals only. Meteorological studies from Tilgner et al. 2014, revealed a more stable stratification during this event with no large differences observed in the coefficient of divergence of the particles, thereby suggesting that the possibility of entrainment of polluted air masses from above was rather low. The increased trace metals are often found from traffic emissions relating to car brake wear and/or fuel combustion. The GB measurement site was close to the main road linking GB and Mt. Schmücke and could be influenced locally by the traffic especially during weekends (period of this event) when more touristic activities are present in this region. This, thus, suggests that traffic emissions may have influenced the concentrations of these trace metals during this event.

P.10, lines 1-13: It is not very clear what statement is related to what sample. Please

improve.

- This section has been improved, and paragraphing has been implemented to separate the various comments. In addition, the statements referring to specific samples have been linked.

p. 10, line 3-5: It is not clear if the words "in general" are related to selected 4 cases or much broader number of events.

- The word in general has been removed, since it led to ambiguity.

Fig 2 and 3: Graphs contain 5 points, but only 4 events are discussed.

- Figures have been updated.

Chapter 3.2.1. : It is difficult to speculate about sources based on 4 data points. The only important fact seems to be higher decrease of levoglucosane concentration in comparison with these four combustion related metals (K, Zn, As, Pb). The crustal element correlation with OC shows, as mentioned in the text later, that the basic correlation among aerosol components is often driven by meteorology. The same probably holds for fig. 4.

- The suggestion with respect to the sources has been considered and the section has been revised accordingly. The speculation to sources has also been weakened. Pages 11, lines 6-30. This section now reads:

Nevertheless, since these correlations are based on statistically few data points their values are used to mainly indicate possible trends between these components. In some cases these trends match observations observed elsewhere. Alongside potassium, levoglucosan is known (Schkolnik and Rudich, 2006; Simoneit et al., 1999; Iinuma et al., 2007) to be a good tracer for particles originating from biomass burning. The correlations with levoglucosan could be indicative of a common source, such as biomass burning. Zn amongst other elements has been observed in biomass combustion emissions (Fine et al., 2004) and although it is not largely accumulated in plants as has been

observed by Schmidl et al.(2008), plumes of biomass burning as well as the applied fuel (Boman et al., 2006) or emissions from the walls of the combustion chambers can influence their concentrations in the atmosphere. These correlations, however, may also be simply due to changes in similar meteorological conditions. Table 2 shows the correlation coefficients of levoglucosan and other trace metals in PM1.2 particles at both valley stations before and after the passage of air mass through the cloud. Positive correlations were also observed for Mn and Fe with levoglucosan at Goldlauter the upwind side. However, changes in the correlations were observed as the air mass passed through the cloud at the mountain station before arriving at Gehlberg. As shown in Table 2, the correlations of K, Fe, and As with levoglucosan became weaker suggesting that deposition of levoglucosan and particles containing such metals during the air mass transport over the mountain to GB may have significantly affected the state at which these components were present at GL. Levoglucosan is a water soluble compound that can dissolve in cloud water and may also be lost from particles, aqueous reaction in cloud drops, or deposition of the cloud droplets. Such losses may account for the lower (about 40%) concentration of levoglucosan observed at the downwind stations as depicted in Figure 2b and the observed change in the correlation with the trace metals observed in Table 2.

In addition the statements that suggested similar source regions based on the correlations have been removed, i.e. Page 12, 28-29 and Page 13 lines 3 – 10.

Chapter 3.2.2. : The same is valid as for previous chapter and correlations. Discussion of sources is quite speculative based on 4 points. The authors are adviced to study them only from point of view of their changes between the sites.

- Same as above, section has been revised and the discussion with respect to the sources has been reviewed and arguments weakened on Page 13 lines 6 – 19. The text now reads:

“Good correlations were also observed for selenium with EC as well as Zn with OC.

As explained above changes in meteorological conditions may lead to correlations between aerosol components. However, it has been observed elsewhere that these components could originate from sources such as fossil fuel or coal combustion (Hashimoto et al., 1970; Pacyna, 1984; Zhang et al., 2014), long range transport or traffic emissions relating to tire or brake wear (Handler et al., 2008).

p. 17, line 21. Probably should be Fe (III) instead of Fe (II):

- Fe (II) has been replaced by Fe (III)

p. 17, line 25. The sentence needs correction.

- The sentence has been rephrased. And now reads “Due to the fast oxidation of Cu (I) to Cu (II) it is easier to measure Cu (II) than Cu (I).”

p. 18, line 17. Table numbers should be corrected.

- Table numbers have been corrected

p. 20, line 18-20: Remove the sentence, it partially contradicts the next sentence and there is no need to speculate about aerosol origin in this paper.

- The Sentence has been removed.

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