Atmos. Chem. Phys. Discuss., 11, C7842–C7849, 2011
www.atmos-chem-phys-discuss.net/11/C7842/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol particle number size distributions and particulate light absorption at the ZOTTO tall tower (Siberia), 2006–2009" *by* J. Heintzenberg et al.

J. Heintzenberg et al.

birmili@tropos.de

Received and published: 16 August 2011

Your comments appear in normal print, our replies appear in italic face.

p. 1155 line 3: Why the authors only mention aerosol formation observed "over other boreal forests"? (...)

According to your suggestion, we have greatly expanded the introduction and now include a more thorough motivation as well as a comprehensive review of the available literature on aerosol measurements over the Siberian forest, including, e.g., Koutsenogii and Jaenicke (1994), Vartiainen et al. (2007), and Paris et al. (2009).

What was the upper size-cut in DMPS and PSAP measurements? What was (were) the CO sampling height(s)?

Calculations of gravitational settling in the horizontal section of the sampling line (10 m) yielded a 50 % transmission probability for particles around 3 μm. This can therefore be considered as something like the upper cut-off size. CO was sampled at 50 and 300 m. With few exceptions, there is no significant difference between the mixing ratios at the two heights. The data shown in Fig. 4 are averages of the two heights.

p. 1162 line 3: Is May already a summer month in Siberia? Any reference to meteorological data?

Some climate discussion says so but to avoid confusion we eliminated the summer and winter terms from the criticized sentence.

Lines 8-9 (and in conclusions): This is most probably true but natural (biogenic) aerosol sources might as well explain a part of the higher concentrations. Which types of sites are referred with "pristine"?

Since this term is hard to define, and to avoid confusion we eliminated the reference to "pristine" in the paper..

Lines 12-13: The sentence "This suggests that..." is a very general statement and I'm not sure what its meaning is.

The text was reformulated as "For all integral parameters and all seasons the concentrations are higher at 50 m than at 300 m, with their ratio being highest in the number (ca. 1.3) and lowest in the volume concentrations (ca. 1.1). This suggests the presence of particle number sources near the ground."

Line 14: It might be relevant to mention that the aerosol volume here includes only the volume of smaller than 835 nm particles. While it's well known that boreal forests are an important source of particles, also in mass-wise (e.g. Tunved et al., 2006; Koutsenogii et al., 1997), the DMPS-derived volume might differ from the total aerosol

ACPD

11, C7842–C7849, 2011

volume significantly, especially in summer.

Thank you. We added sentences to the manuscript stating the upper size cut (equals roughly PM1), coarse particles as a possible source of aerosols in summer. Also, the work by Tunved et al. (2006) is now acknowledged.

p. 1163 line 5: What here means “evens out”?

It means the curve becomes more even and less structured. The revised text now avoids this term.

line 14: Is the measured aerosol particle diameter range the same both for the absorption coefficient and for the volume?

Yes, indeed. All instruments sample from the same inlets.

p. 1165 line 11: I don’t see a clear late winter maximum in mode 3.

Sorry about the typo related to modes 2 and 3. The winter maximum pertains to mode 2.

p. 1166 line 22-23: Were the trajectories, computed for the different heights, generally consistent with each other?

Yes, quite so, probably because of the homogeneous topography of the larger Siberian forest region. See also our trajectory study in Heintzenberg et al. (2008).

p. 1170: When discussing the clusters authors might (whenever possible) refer to the air mass types instead of referring to the cluster numbers (which seem to be in arbitrary order, devoid of any information on the air mass origin or type).

Thank you, this feature has now completely been revised in the text, Table 2, and the Figures 8-10. In particular, the clusters were sorted according to their seasonality. Cluster 1 contains mainly mid-winter periods, and cluster 10 mainly mid-summer periods.

ACPD

11, C7842–C7849, 2011

Line 20: The particle mass concentrations are presented here for the first time. How these were derived?

Particle mass concentrations were derived from number size distributions assuming spherical particles and a gravimetric density of 1.6 g m^{-3}. This gravimetric density usually reconciles calculated an measured fine particle mass in closure studies with Central European tropospheric background aerosols.

p. 1171 lines 4-7: This is confusing to me. I don't find real proofs for claiming that biomass burning and new particle formation do not greatly influence the mean concentrations, even though their effect is undetectable in back-trajectory analysis. First of all, what then influenced the concentrations, anthropogenic sources?

Sorry for the misunderstanding. We did not negate that there are several options for natural aerosol formation, but feel that the main reason for the misunderstanding was that the text has been compiled in an unfortunate fashion.

Second of all, I think these issues should be inspected more carefully by separate papers (as the authors have promised to do in conclusions) using the available modelling and satellite tools.

We feel the same and have a careful inspection of satellite images in mind (e.g., MODIS), which are helpful in detecting plumes of forest fire smoke. Meanwhile, this seemed out of scope for this article.

On the other hand, if the purpose of these sentences is to say that the concentrations remained relatively low at all clusters (except for those two anthropogenically influenced from south) it seems slightly contradictory to: 1) first say that the mean concentrations were higher than those at truly pristine sites indicating anthropogenic influences, and 2) then to say that biomass burning or particle formation didn't increase the mean concentrations because they were so low. Also in sections 3.1.1 and 3.1.2 forest fires and particle formation are suggested to explain many of the observed

ACPD

11, C7842–C7849, 2011

seasonal characteristics of concentrations and modal parameters. In the end, I find it difficult to figure out what the authors suggest the relative contribution of different sources (anthropogenic vs. natural) is, even though there is a considerable discussion on the possible sources in this paper.

Again, we feel that indeed our text has been misleading. We acknowledge the presence of the following particle sources for fine particle mass: biomass burning (mainly in summer), secondary formation (mainly in summer, from biogenic precursors), anthropogenic emissions (most effective in winter). To more efficiently combine the discussion based on the results in sections 3.1 and 3.2 we compiled the discussion in a more stringent manner in a new discussion section 3.3. Meanwhile we also outline why we have difficulties in attributing particular fractions of the observed particles to particular sources: "Although we found clear indications for the effects of specific groups of particle sources in the aerosol data, we are currently unable to quantify their possible contribution to particle number and mass. A main reason is the lack of source-specific aerosol information, such as chemical composition. Our analysis is also hampered by the lack of continuous local micrometeorological measurements for the period 2006-2008, such as 3d wind on the tower. Such data, which is in process of being collected at ZOTTO, can help clarify the possible role different aerosol particle sources and also vertical atmospheric exchange.

In addition, I miss some more discussion on the observed similarities and/or differences between the results of this study and those reporting previous findings from Siberia (which though are not so many). At least the findings of those articles cited (Paris et al., Koutsenogii et al., Vartiainen et al., Kozlov et al.) could be touched. Now comparison is made only with number concentrations found in Vartiainen et al.

We now include a more comprehensive comparison of our observed concentration data with those in the mentioned works.

Comments on Tables and Figures: Table 2: Average total concentrations here (be-

ACPD

11, C7842–C7849, 2011

tween hours 14 and 20) are much lower than those medians given in Table 1. Is this truly explained by the diurnal variation? Could the authors comment/present how pronounced the observed diurnal variation was? The boundary layer diurnal evolution and structure are broadly discussed in the paper so it might also be convenient to discuss a bit on its effects on the observed concentrations in different seasons (and heights).

Thank you for pointing out this inconsistency, which made us discover an error (taking natural instead of base10 logarithms) in our size integrations. Table 2 and related figures have been revised accordingly.

Fig 3: Instead of "sap" I would use "sigma ap" also in axis and figure legend. Fig 4: Same as for Fig. 3, I might use "sigma ap" instead of "sap".

Done.

What was the diameter range for volume and absorption?

The limiting diameter for volume was 835 nm, the upper cut-off size for the absorption measurement roughly 3 μm, defined by the inlet characteristics.

Fig 5: I don't understand how the probability lines can be continuous?

The lognormal fitting algorithm allows for all values of DG between minimum and maximum. The actual fitting results were classified for a histogram PDF in 14 logarithmically even diameter classes between 10 and 1000 nm. The number of cases in these 14 classes was divided by the total number of cases. In the plot we allowed Excel to smooth the lines.

Figs 8 and 9: It's difficult to separate between some of the darkest colours. The numbering should follow some logic, e.g. numbers from one to ten could follow from instable to stable profile, respectively. This would also help in reading the text.

Thank you. We found it difficult to choose colors that bear no possibility at all of being mixed up. Regarding the numbering feature, this has now completely been revised. In

ACPD

11, C7842–C7849, 2011

particular, the clusters were sorted according to their seasonality. Cluster 1 contains mainly mid-winter periods, and cluster 10 mainly mid-summer periods.

In addition, for month axis (Figs. 2-4, 7) a scale from 1 to 13 would be clearer to me.

This seems to be a matter of personal taste. To us it makes most sense that the left end of each column marks the start and the right end the end of the respective month.

p. 1157 line 2: Check the given diameter range (12-835 nm). In next page, a bin range from 15 to 835 nm is mentioned.

Thanks for the hint. The numbers were correct but the reduction in size range was unexplained.

Several technical remarks: p. 1154 line 14: considerably; p. 1156 line 3: The scientific leaders of the ZOTTO facility are..., or rephrase and name only one leader ...together with...; p. 1162 line 14: "in summer" is repeated; line 15: than; p. 1164 line 18: four (?) summer months; line 21: "in summer" repeated; lines 22-24: a bit complex sentence, consider rephrasing; p. 1165 line 1: considerably; p. 1172 line 10: considerably;

Changed, thank you.

p. 1166 line 8: In the presence...

Sorry, this appears to be a misunderstanding. We changed the sentence to make the absence clearer.

p. 1167 line 17: could the cluster number "k" be marked with a different symbol to separate between it and the distance factor k_i.

Thank you, the distance factor is now labeled as l_i.

p. 1170 line 12: I assume this should be "Figure 10", which is missing

Sorry, that figure had disappeared during the ACP editing but has been added since.

p. 1171 line 10: "ZOTTO exhibits only a limited intensity of aerosol sources on its own."

ACPD

11, C7842–C7849, 2011

This sentence was indeed misleading, and consequently omitted in the revised version. Instead, we added a comprehensive discussion on the possible sources in a new section 3.3.

Wolfram Birmili, on behalf of all co-authors.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 1153, 2011.