Atmos. Chem. Phys. Discuss., 11, C7852–C7860, 2011
www.atmos-chem-phys-discuss.net/11/C7852/2011/

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.

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Received and published: 16 August 2011

Thank you for your numerous comments. We think that we were able to positively consider the overwhelming number of your suggestions to amend the article.

In the following, your comments appear in normal print, our replies appear in italic face.

page 1160: It's not clear to me at which height the TSI nephelometer was measuring; closer to the 50 or 300 m?

In section 2.4 we stated that the nephelometer also samples both inlets, and how its data were used.

I found some inconsistency regarding the origin (sources) of the measured particles: p 1155, l 6 “At this site, anthropogenic influences are of minor importance, so that the sampled aerosol can be representative for a very large spatial area.”

The sentence was meant to say that the ZOTTO site is far from any substantial settlement or large-scale industrial plant.

p 1162; l 8-10: “They are higher, however, than the concentrations at truly pristine sites (Andreae, 2009), indicating a contribution from anthropogenic sources.” (Also in conclusion.)

Since it is difficult to define this term, and since it is questionable how many areas in Siberia are really still “truly pristine”, we eliminated the confusing reference to “pristine”.

p 1162: l 12-13: “This suggests the measurement at 50 m to be influenced in a stronger fashion by particle sources”.

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.” Meanwhile we acknowledge that “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” [process nature as well as] “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, could clarify the possible role different aerosol particle sources and also vertical atmospheric exchange.”

p 1171 l 8-10: “The results underline the location of ZOTTO ... apparently exhibits only a limited intensity of aerosol sources on its own.” For me, these statements give

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a confusing impression. On the one hand it's stated that local sources are of minor importance, but on the other hand the enhancement of concentrations at a lower lever seem to indicate a surface source somewhat near the measurement site. Also, is the contribution of anthropogenic sources deduced from anything than the higher concentration, by elimination of other explanations? For example, the mean concentrations reported here fall in the range reported eg. Dal Maso et al (Tellus, 2008, 60B, 495-508) or Tunved et al (2005, JGR 110, D07201, doi:10.1029/2004JD005085) for various boreal forest sites, with variable influence of possible natural and anthropogenic sources. It is not clear what is meant by 'truly pristine'. I think a more clear discussion of what the authors mean by sources could be given: near/far, natural primary/direct human emission/secondary natural/secondary anthropogenic? The trajectory clustering analysis is well made and I think the results are valuable. The presentation of the results and the interpretation could be made somewhat clearer, as at least for me following the numbering system and trying to connect it to specific directions was quite some work.

Thank you for your detailed comments. In fact, we confirm that our previous discussion was somewhat contradictory. With respect to the question of local sources near the tower, we now wrote in Section 3.1: "Local particle sources, which include the diesel power generator of the station and sporadic truck traffic to the village of Zotino are unlikely to cause this discrepancy. A reason is that the signature particle size distribution effects that are typical for diesel engine emissions have neither been encountered at the 300 m nor at the 50 m level."

Further, since the vertical gradient is more sensitive to particle number, one can therefore mainly think of a number-relevant particle source, which includes secondary formation. In general, we truly 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 now compile the discussion in a more stringent manner in a new discussion

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section 3.3. Some extracts from this new text read as follows:

3.3. Discussion

The results are now discussed with respect to their implications on the possible aerosol sources over Siberia. We presume that the ZOTTO aerosol measurements at the 300 m level is likely to be influenced by the following types of aerosol sources: 1) biomass burning, i.e. natural as well as man-made forest fires, 2) secondary formation, generating fresh particles as well as additional particle mass from gas-to-particle conversion, 3) significant anthropogenic point sources, such as industrial plants, 4) diffuse anthropogenic sources, such as domestic heating by private households, motor and ship traffic. In addition, aerosol from outside, both natural and anthropogenic can be transported into Siberia through horizontal advection.

3.3.1. Biomass burning

In the case of CO, the impact of biomass burning was clearly detectable during the warm seasons 2007 and 2008 (Vasileva et al., 2011). Our aerosol data revealed a significant annual peak in the month of July (Fig. 2). A visual screening of the particle number size distributions for individual events suggested that on 19/07/2007, ZOTTO was remarkably influenced by forest fires within a range of a few 100 km. This event could be recognized by a strongly time-dependent signal of accumulation mode particles. However, if forest fires occurred farther upwind, the air mass would attain a higher spatial homogeneity so that it would be difficult to identify such events from particle size distributions alone. Further evidence on this was provided by the ratio sigma-ap/V, which serves as a proxy for the mass-specific light absorbance of the particle material. This ratio attained an absolute minimum in July as well (Fig. 3). Kozlov et al. (2008) showed that the mass fraction of black carbon in sub-micrometer aerosol can serve as an indicator

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of influence of smoke from remote forest fires in Siberia. Their long-term study revealed that this fraction in smoke of remote forest fires is lower than in the background aerosol, i.e. fresh biomass burning smoke is less absorbing. The observation in Fig. 3 is therefore in line with the maximum in particle number and volume concentration in July, when there is a maximum likelihood of forest fires. Although we conclude that forest fires are the likely cause of the July concentration maximum, it is difficult to judge their quantitative contribution, left alone .

3.3.2. Secondary formation

In boreal forest environments, the formation of new atmospheric particles (diameter $<$30 nm) has been found to occur most frequently in spring (dal Maso et al., 2005, 2008a). Figure 2 shows elevated particle number concentrations (relative to particle volume) in the months February-April and June. At least for the month of March and later, it is plausible that secondary particle formation influenced the measurements at ZOTTO. A preliminary visual screening of the time evolution of the particle number size distributions suggested the occurrence of 10-20 particle formation and growth events per warm season, depending on the exact numerical criterion used. Fresh particle formation seems, however, not to occur often enough to show up in any of the trajectory cluster size distributions (Fig. 10). In fact, numerous days showed characteristics such as a nucleation mode diameter considerably bigger than 30 nm, which suggest particle formation not near the site, but some way upwind. Although we refrain from a detailed analysis at this stage, the frequency of fresh particle formation events appears in any case to be substantially lower than at continental sites in Finland (dal Maso et al., 2005, 2008a) or Germany (Hamed et al., 2010). It is not clear whether it is a lack of certain necessary gaseous precursors or particular meteorological circumstances that make particle formation and growth events less

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visible in the ZOTTO data. Apart from the atmospheric particle number budget, secondary formation over boreal forests is, in any case, expected to have a significant impact on particle mass (Tunved et al., 2006). The minimum particulate volume V at ZOTTO was observed in October (Fig. 2). During the warm period (March-September), considerably higher V were observed. While this observation could give a hint to secondary mass production, such as from biogenic precursors, the corresponding contributions are hard to distinguish from a possible background from biomass burning contributions.

3.3.3. Anthropogenic sources

Some of the strongest variation in the data set includes the annual cycle of particle volume V (Fig. 2), the ratio sigma-ap/V (Fig. 3), and also the variation of the number size distribution as a function of season and air masses (Fig. 10). It is a plausible conclusion that the winter maxima in V and sigma-ap/V (a measure for the mass specific absorbance) are related to anthropogenic fossil fuel combustion. The most significant single dependency adds to this impression: trajectories from latitudes far south of 60 deg N are associated with the highest concentrations of particle number, volume, and also CO (cf. clusters No. 2 and 5 in Fig. 8 and Tab. 2). Fig. 8 illustrates that the Siberian centres of population and industry concentrate on a belt around 55 deg N, and are located south-west to the ZOTTO site. As can be grasped from Table 2 and Fig. 10, the aspect of geographic origin is of at least equal importance to the aspect of seasonality, since cluster No. 5 - associated the by far highest particle concentrations, shows only a relatively modest seasonality towards the winter period (season index +0.3). A closer look at several individual case studies confirmed the large-scale nature of the anthropogenic effect. Polluted air masses often arrived when the back trajectories switched to south-westerly directions, and such situa-

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tions often remained stable for a few days. The temporal continuity of the size distributions in many such cases suggested that the polluted air mass had achieved a high spatial homogeneity during the transport from the population and industrial centres to the remote ZOTTO site. This also weakens the possibility that the mean size distribution ZOTTO is influenced by the few local anthropogenic sources within a few 10 km.

3.3.4. Limitations

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, could clarify the possible role different aerosol particle sources and vertical atmospheric exchange. We are also aware that a more detailed classification and analysis of the time series might provide additional hints to the magnitude of particular aerosol sources.

Some suggestions: - label each cluster according to the 'name' given in Table 2 in Figs 9 and 10. The number could also be given for easier comparison to Fig. 8. Thank you, we added such labels, and also renumbered the clusters according to the criterion of vertical stability and/or wind direction.

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.

- In the caption of Fig 9 it is said that the theta v profiles allow for a clear distinction

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between more and less stable stratification. This information could be added to Table 2.

Thank you, done.

- A suggestion for clarifying the message about "the slower the air mass and the more it originates from the southerly latitudes, the higher the mass and CO": this could be seen clearly in a figure with mass/CO plotted versus parameters like the 144 h trajectory length and for the 'southness' maybe the average latitude of the trajectory. This is most certainly not something that I would expect to be included in the MS, but is something that I personally would try to do.

Thank you for this suggestion. For a future paper, we have a careful inspection of satellite images in mind (e.g., MODIS), which are helpful in detecting plumes of forest fire smoke. In this context, the 'southness' aspect could be analysed in an attempt to more clearly separate the contributions from biomass burning and anthropogenic combustion (mainly fossil fuel). Meanwhile, this seemed out of scope for this article.

On tables and figures: Table 2: Regarding the summer/winter parameter, with what criterium is a cluster called summer or winter? Eg. cluster 3 has no season identifier with an absolute s/w value of .3, but cluster 1 is summer with the same value. Also, clusters 4 and 6 have opposite season descriptors but both have quite high positive s/w-indexes (typo?).

Sorry for the mistake, cluster 4 should be dubbed "Central Europe, winter". Clusters with an absolute s/w value of 0.3 or less are not assigned a season identifier. An exception is cluster 1, since it has almost the same trajectory as cluster 10, but contrasts in season with that cluster. Seasonal information has now also been added to Table 2 and Figures 8-10.

I don't understand fig. 5. To my understanding, integrating the whole pdf over the Dg space should give 1 as a result. This might be so, but the way of plotting it leaves it

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unclear whether the integration should be performed in log space or in linear Dg space. Depicting the pdf in a log scale in the latter case seems quite confusing in my opinion, as the area under the curve does not correspond to the actual probability of finding a given Dg range.

The lognormal fitting algorithm allows for all values of DG between minimum and maximum. The actual fitting results were classified for a histogram-type 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.

In figure 6, for completeness, I suggest that also a median size distribution for eg. the particle number concentration percentiles from eg. 47.5-52.5 (5% around the median) could be given; I guess that these concentrations are the most frequently observed and thus representative for the 'general' aerosol.

We added the suggested curve to Fig. 6, thank you.

Technical: Figures: In some figures, the sigma ap in the legend has turned into an s.

Changed to a consistent sigma ap in legends and figures.

p. 1166 l 8: Missing 'In'

Added, thank you.

Wolfram Birmili, on behalf of all co-authors.

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