Interactive comment on “In situ measurements of aerosols optical properties and number size distributions in a subarctic coastal region of Norway” by S. Mogo et al.

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First of all, we would like to thank referee #2 for his constructive comments and suggestions. In the following the comments will be addressed and discussed.

General comments

1. Make it clear in the text what are the differences between the results presented in this work and Montilla et al. (2011), Rodriguez et al. (2011). Make additional references to the findings of these papers in the text and compare to their results.

The paper by Montilla et al. (2011) presents a new instrument (the IS3) developed to measure continuous spectra of aerosol absorption coefficients. The IS3 technique had been used in the same field study that we are describing in the present work and an intercomparison of data between the PSAP and the IS3 is presented by Montilla et al. (2011) for laboratory generated aerosols and for the ambient aerosols observed in ALOMAR during this campaign. This is made clear in the manuscript, as explained in our response to the 5th specific comment from the reviewer. However, the IS3 instrument is based on collection of aerosols in filters and due to weather conditions, not all the the days were available for Montilla et al. (2011) to compare with the PSAP and consequently, less data is presented on that work.

The paper by Rodríguez et al. (2011) is dedicated to the columnar properties of the aerosols and the data presented was obtained from a Cimel sun-photometer since 2002 until 2010. We reference this work because, the Cimel instrument provided the information necessary to identify possible dust events with a methodology that is now explained on the manuscript (by request of reviewer #1). Also the work presented by Rodríguez et al. (2011) conducted a full study on back trajectories in Andenes and we used their classification in our work. However, no comparison of the in situ measurements with the Cimel’s column data was performed because there is a strong limitation to this comparison in our field study due to the short term of the campaign. Actually, the comparison of the surface in situ measurements with the columnar data is always a complicated issue even in more favorable conditions of measurement. But in the ALOMAR station the problem is intensified by the strongly limited number of days with available columnar data due to cloudiness over the region. This situation leaves only a few days with data available from the Cimel instrument and no significance can be attributed to the results. Because of this reason, the usefulness of the columnar data becomes restricted to punctual events that occur in days with clear sky. This explanation was also presented to reviewer #1, who asked for comparison of in situ versus...
columnar data to indicate how representative the surface measurements are of the atmospheric column.

2. Shorten the text and reduce the number of figures whilst retaining the important messages coming out of the analysis of the datasets. Some of the figures and very detailed discussion could be moved into Supplementary Material. I strongly recommend combining the air mass origin analysis with the discussion about optical properties and size distributions (e.g. move section 3.4 to beginning of section 3 and combine with section 3.1). It would also be more informative to show particular examples (events) of air masses containing different aerosol types such as dust aerosols (where do they come from?) or continental aerosols (be more precise about what this means). This would avoid many of the rather vague statements in section 3. Also, are there no differences between continental aerosols coming from sectors 1, 2 or 3? The types of possible “continental” aerosols should be discussed in more detail.

The text has been improved in several points, according to the suggestions of all the reviewers as detailed in the respective specific comments.

Figure 7 is now included as Supplementary Material. Several other figures were grouped or combined by suggestion of reviewer #3. The manuscript has now 8 figures, instead of 13 in the previous version.

The suggestion about combining the air mass origin analysis with the discussion about optical properties and size distributions is accepted and the manuscript changed accordingly as detailed in the specific comments. Some particular events are briefly presented.

Regarding to the types of continental aerosols arriving from sectors 1, 2 and 3, they are hard to differentiate from each other because the air masses arriving from these sectors are the less frequent during our study and only a few cases of each type were registered. See the frequency histogram, Fig. 8 (old Fig. 12). However, with the new structure of the text, combining the analysis of air masses origin with our data and the example situations presented, we think that the possible differences between these particle types became apparent. Aerosols arriving from sector 2 stands out most, with higher optical parameters and higher number of particles in the micrometric range but lower number of particles in the submicrometric range. Sector 2 also presents the most outstanding size distribution, with a monomodal shape and presenting a peak near 100 nm.

3. The abstract and conclusions need to be re–written so that the main findings are clear to the reader. Rather than lists of numbers, more general characteristics and findings as well as wider implications need to be summarised. 

Done. The abstract and the conclusions have been improved as suggested.

Specific comments

• Page 32924, lines 20–25: refer to more recent references on soot deposition on snow.

We added 4 recent references and the text is changed to: “... In addition, the deposition of light-absorbing particles onto snow and ice reduces the surface albedo, which, in turn, affects the snow pack and the Earth's total albedo (Law and Stohl, 2007; IPCC, 2007). A recent study (Hadley and Kirchstetter, 2012) generated and characterized pure and black carbon-laden snow in the laboratory and verified that black carbon contamination appreciably reduces the snow albedo, at levels that have been found in natural settings (Forsström et al., 2009; Aamaas et al., 2011). Clarke and Noone (1985) found that for a mass fraction of 10–40 ng g\(^{-1}\) soot, the snow albedo is reduced by 1–3 % in fresh snow and by a factor of 3 as the snow ages and the light-absorbing particles become more concentrated. Doherty et al. (2010) estimated the black carbon concentrations in snow for regions near the Arctic at 4–20 ng g\(^{-1}\), with an expected albedo reduction of 1–2 %. ...”
• Page 32926, line 26: it would be useful to include the paper layout here.
  Done.

• Page 32929: lines 10–13: explain why the gap in number size cannot be bridged. The gap between the size ranges is not that large and it would be possible to interpolate (as pointed by reviewer #3) but the problem here is the low concentration of particles registered by the APS instrument. With such small concentrations, the error resulting from interpolation and conversion of diameters isn’t advantageous. This is now clarified in the manuscript: "... This decision, together with the low concentration of particles registered by the APS, prevented us from merging the SMPS and APS data because the gap between the upper channel of the SMPS, 390 nm, and the lower channel of the APS, 500 nm, would require interpolation between these sizes and a conversion of the aerodynamic diameters provided by the APS to the mobility diameters provided by the SMPS, with associated errors resulting from both procedures”

• Page 32932, line 6: define P25.
  Done. P25 is the first quartile, it is now defined in the text.

• Page 32932, line 15: yes, but what do Montilla et al. (2011) show?
  This item is already explained in the first general comment of this reviewer and in the manuscript we explained that: "Montilla et al. (2011), measured continuous spectra of aerosol absorption coefficients in the wavelength range of 320–800 nm and obtained the αa exponents for different spectral ranges, including the UV range.”

• Page 32933, lines 5–14: this is an example of where it would be better to discuss these points together with the back trajectory analysis and highlight particular events.
  Done. We restructured the manuscript by creating a subsection 2.4 in the methodology section, where we explain the procedure for analysis of the transport pathways of the air masses arriving at ALOMAR. The discussion of the optical parameters is now linked with the study of the back trajectories.

• Page 32924, lines 20–29: likewise, link to back trajectory analysis.
  Done. We combined the size distributions and the back trajectory analysis.

• Page 32936, lines 2–6: it is unclear what is really meant by “northern European aerosol”? Are there no differences between coastal sites like Alomar and continental sites like Pallas? (biogenics, sea–salt etc.)?
  The classification “northern European aerosol” is based on the criteria of Asmi et al. (2011) and refers to stations similar enough to be described in the same type, dominated by clean continental and atlantic aerosol but also with occasional influence of more polluted air masses. We included ALOMAR in this group because the bimodal shape of the median size distribution, the concentrations of the particles in each size range and the shape of the area with greatest density of points in the scatter plot of \( N_{30} \) versus \( N_{100} \), all yield similarities with those characteristics found for the group of “northern European aerosol” by Asmi et al. (2011). However, the reviewer is right, ALOMAR and these stations (Birkenes, Vavilhill, Aspvreten, Hyytiälä and Pallas) present similarities but also each one presents its own characteristics. ALOMAR presents an intermediate behavior between Birkenes and Pallas, with the concentration of particles lower than in Birkenes but higher than in Pallas. The shape of the median size distribution is more similar to the shape registered in Birkenes, with the bimodal aspect more clearly defined than in Pallas. On the contrary, the shape of the area with greatest density of points in the scatter plot of \( N_{30} \) versus \( N_{100} \) is more similar to that of Pallas, suggesting cleaner air from the Arctic or Atlantic Oceans, than that on Birkenes. This discussion is now included in Section 3.2.

• Figure 8: in the text the implications of what is shown for Figs.9 and 10 need to be described more clearly.
Fig. 9 is now the Fig. 5e,f of the new manuscript and is described as follows:

"Fig. 5e illustrates the relationship between the scattering and absorption coefficients. This represents another way to analyze the single-scattering albedo parameter and the low values of $\sigma_s$ compared to those of $\sigma_a$ are responsible for the high $\omega_0$ values observed. In Fig. 5f, the relationship between the Angström exponents is also presented, and two regions can be identified as exhibiting a higher density of data. Region A, with higher $\alpha_s$ and $\alpha_a$ values, indicates light absorption by smaller particles, and region B, with lower $\alpha_s$ and $\alpha_a$ values, indicates light absorption by larger particles. These two regions represent the two modes that were observed in the frequency histogram of the $\alpha_s$ parameter (Fig. 3d). In Fig. 3d, the higher density near $\alpha_s = 0.7$ and $\alpha_s = 1.9$ and the lower density near $\alpha_s = 1.3$ should be noted. The $\alpha_s$ values can be explained by the aerosol size distributions measured by the APS, as the count mean diameter of the size distribution (see Fig. 7c and its explanation in the text) presents a correlation of $-0.70$, with smaller particles associated with higher $\alpha_s$ values. Because $\alpha_s$ depends on the chemical composition of the particles (as mentioned before, different aerosol species present different $\lambda$ dependencies on light absorption), we conclude that region A, which has higher exponents due to the presence of smaller particles, presents the characteristics of continental polluted aerosols, which may originate from continental urban sources. Region B, which has lower exponents due to the presence of coarse particles, which are clean and less absorbent, may be of marine origin."

Fig. 10 is now the Fig. 6 and it is also more deeply explained as follows: "Fig. 6 displays the value of $\omega_0$ as a function of the scattering/absorption coefficients and the Angström exponents. A comparison of $\omega_0$ with $\sigma_s$ and $\sigma_a$ was conducted to identify the darkest and the whitest aerosols observed. The data are classified according to $\sigma_s$ into three groups, namely $< 0.23$ Mm$^{-1}$, $0.23 - 0.48$ Mm$^{-1}$ and $> 0.23$ Mm$^{-1}$ (corresponding to the first quartile, second + third quartiles and fourth quartile, respectively). Any absorption value may lead to the lowest values of $\omega_0$, whereas for scattering, the lowest $\omega_0$ values are observed in the lowest $\sigma_s$ range ($< 5$ Mm$^{-1}$). A comparison of $\omega_0$ with $\alpha_s$, shown in Fig. 6c, was conducted because the wavelength dependency of the absorption is related to the composition of the aerosols. The most absorbent particles exhibit $\alpha_s$ values below 0.6, while the less absorbent particles present higher $\alpha_s$ values, reaching near unity. In Fig. 6d, $\omega_0$ is also compared to $\alpha_s$ to search for information on the size of the aerosols and to examine whether the optical data can provide support for the size distribution measurements. The joint analysis of Fig. 6a and Fig. 6c shows that, for a given $\sigma_s$ value, lower $\omega_0$ values correspond to higher $\alpha_s$ values, and higher $\alpha_s$ values correspond to smaller $\alpha_s$ values. This result agrees with that obtained by Clarke et al. (2007) and, considering that $\alpha_s$ is higher for smaller particles (see Fig. 7c and its explanation in the text), one can conclude that, for a given $\sigma_s$ value, lower $\omega_0$ values correspond to smaller particles, and higher $\omega_0$ values correspond to larger particles. In Fig. 6e, the single-scattering albedo, $\omega_0$, is plotted against its own exponent, $\alpha_{\omega_0}$. The spectral shape decreases mainly with the wavelength, $\alpha_{\omega_0} > 0$, but in some cases, the single scattering albedo increases with the wavelength ($\alpha_{\omega_0} < 0$). This situation has been reported to be a result of the arrival of dust (Rodríguez et al., 2011). Only a few such occurrences took place during our measurements, and these cases were characterized by higher total particle number concentrations, such as 3919 cm$^{-3}$.

According to Cappa et al. (2009), the relationship between $\omega_0$, an intensive aerosol optical property, and $\sigma_s$, an extensive property, can be used to differentiate between background aerosols and the inputs of primary aerosols. Applying this methodology at the ALOMAR station, we observe predominantly high values of $\omega_0$ caused by the very low $\sigma_s$ values. This fact, together with the $\alpha_s$ values registered, allows us to describe the local air as extremely clean and only episodically influenced by small particles resulting from long-range transport. This conclusion is also supported by the results of other techniques, such as the analysis of the air mass origins (see Sect. 3.4) and the use of MODIS images and columna
• Figure 12: this figure is confusing and needs to be simplified. Combine with information presented in time series in section 3.1. All the information and interpretation relative to this figure is now connected with the time series of the optical parameters and the size distributions. The figure itself was improved and we think it is more legible now.

• Acknowledgements: It is probably a question of wording but it sounds like the U. Helsinki was responsible for POLARCAT activities? The international POLARCAT-IPY activity was coordinated by Andreas Stohl (NILU) and Kathy Law (CNRS) with coordination based at NILU, Oslo, Norway. Corrected to: “... Also, we thank the POLARCAT staff, the Division of Atmospheric Sciences (Helsinki University) team headed by M. Kulmala, and L. Laakso, for calibration facilities.”

References


Interactive comment on Atmos. Chem. Phys. Discuss., 11, 32921, 2011.