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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Regional aerosol optical properties and radiative impact of the extreme smoke event in the European Arctic in spring 2006" *by* C. Lund Myhre et al.

**C. Lund Myhre et al.**

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Anonymous Referee #2 Received and published: 2 August 2007

We thank the reviewer for the thorough and positive review and the time spent to help clarifying the paper with valuable suggestions. All comments are hereby answered one by one:

1) p. 9520, line 14: provide lat and long information for Andenes.

We have rather excluded it from line 6 for Ny-Ålesund to make it consistent with all sites mentioned in the abstract. We think this is to detailed information for the abstract, and the information is already included in Table 1 and referred to in the first paragraph of



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section 2.

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2) p. 9520, lines 15 - 17: Sentence starting with “Importantly, at SvalbardEˇ .” is incomplete and needs to be fixed.

The sentence is rephrased.

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3) The abstract should provide more of a summary of results. The first paragraph does a good job of introducing the topic of the paper but then should go on to report more specifics of what was learned about the evolution of the aerosol optical properties. Also report quantitatively the range of regional radiative forcing values from the smoke periods and compare to background Arctic aerosol.

We have included details about the optical properties as well as the radiative forcing estimates in the abstract, as suggested.

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4) p. 9521, lines 1 - 2: Change to “Eˇ which is a function of their composition, SIZE, shape, and phase.

The sentence is corrected.

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5 ) p. 9521, line 2: Change to “Calculations of the direct effect of aerosols has a high level of uncertainty despite the huge scientific focusEˇ ..”

The sentence is rephrased.

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6) p. 9512, line 7: Provide a brief explanation for the difference in the DRF estimates



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based on satellite observations versus model calculations.

We agree that this is a very crucial and interesting scientific issue. The reason for the discrepancy is uncertain and debated. Thus we think that it is beyond the scope of this work to explain this in more detail, as our point was only to illustrate the uncertainty still existing in the estimations of the direct effect of aerosols. There has so far not been any study reconciling this difference in the DRF estimates, to our knowledge.

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7) Introduction: For clarity, the introduction needs more paragraph breaks separating the topics that are introduced.

I fully agree, and in fact is seems that the breaks were lost during the type set. We have now included breaks at the following locations: page 9521 line 3 page 9522 line 2 page 9522 line 12 page 9523 line 3

_____

8) p. 9522, lines 14 - 17: Explain the connection between highest record temperatures and extensive pollution transport into the region.

As referred to in the paper, this is comprehensively explained in Stohl et al (2007). I have now included the main points this section.

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9) p. 9522, line 15: define PM0.7.

Done

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10) p. 9523, line 3: Should be “Eˇ .source regionsEˇ .”

Corrected



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11) Table 1 and Figure 1: Naming convention for Andoya-ALOMAR-Andenes should be made consistent between the table and the figure.

This is now changed and harmonized.

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12) p. 9525, line 8: Should be “Eˇ based on separate sky radianceEˇ ..”

Corrected

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13) Table 2: Explain the use of a factor of 1.1 to convert from EC to EM. Likewise for the factor of 1.8 to convert from OC to OM. Provide references for these conversion factors.

The argumentation for using a conversion factor of 1.1 for EC is based on the study of Kiss et al. (2002). In an experimental study involving aerosol filter samples at the rural background site K-Puzsta, it was estimated that a factor of 1.1 should be used to a convert EC to EM (Elemental matter). Elemental carbon is usually characterized by a highly condensed aromatic structure with functional groups on the surface. Given one oxygen atom in a polycondensed structure built up of 24 carbon atoms this should correspond to a factor of ˜1.1. If the number of functional groups were higher, then a factor of 1.1 would be a lower estimate.

Reference: Kiss, G., Varga, B., Galambos, I. and Ganszky, I., 2002. Characterization of water-soluble organic matter isolated from atmospheric fine aerosol. Journal of Geophysical Research 107(D21), 8339, doi: 10.1029/2001JD000603.

We have included the reference in the manuscript.



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The conversion factor of 1.8 is adapted by Stohl et al. (2007).

As thermal optical analysis accounts for the carbon content of the organic constituents of the aerosol only, a conversion factor converting levels of OC (μg C m-3) to levels of OM (organic matter) (μg m-3) is needed in order to account for the oxygen, hydrogen, nitrogen and sulphur associated with the molecules. This conversion step is recognized as one of the most important uncertainty factors in mass closure calculations. Turpin and Lim (2001) re-evaluated the commonly used conversion factors applied both for urban and rural areas, ranging from 1.2-1.4 (Gray, 1986). Their study indicates that a ratio of 1.6ś0.2 is a better estimate for urban aerosols. The reason for this increase can be attributed to the increased focus on the highly oxidized secondary organic aerosols, as well as the polyhydroxy compounds associated with primary biological aerosol particles. Furthermore, conversion factors of 1.9-2.3 were suggested for aged aerosols, whereas conversion factors of 2.2-2.6 were suggested for aerosols originating from biomass burning. Typically, the WSOC (water-soluble organic carbon) content tends to dominate the OC fraction of biomass burning aerosols, thus the higher OC:OM ratio used for emissions from biomass burning can be attributed to the highly oxygenated character of these organic molecules. E.g. levoglucosan, which is the most abundant particulate phase component emitted from biomass burning, has an OC:OM ratio of 2.25. On the other hand, emissions from biomass burning also contain a wide range of various methoxy phenols, e.g. guaiacol, which has an OC:OM ratio of 1.5. When attempting to identify the chemical composition of the OC fraction on a molecular basis, one typically fails to account for more than ˜30%, thus arguing for a specific conversion factor without accepting that there is a considerable uncertainty range makes no sense.

An experimentally derived conversion factor of 1.9 for OC to OM was reported by Kiss et al. (2002). To our knowledge, this is the only study reporting a conversion factor for OC that is obtained at a European rural background site based on an experimental



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approach. During the episode described in the present manuscript, a mix of aged aerosols and aerosols originating from wood burning is to be expected, thus using a conversion factor of 1.8 we most likely provide a lower estimate of the OM fraction.

We have performed test calculations to check the effect of changing the conversion factors and conclude that this is of minor importance for the SSA (E.g. an increase in the conversion factor by 30% resulted in a change in SSA of 0.003)

The reference in included in the manuscript.

Turpin, B.J., and Lim, H.-J.: Species contributions to PM2.5 mass concentrations: Revisiting common assumptions for estimating organic mass, Aerosol Sci. Technol., 35, 602–610, 2001.

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14) p. 9527, line 26: What exactly does the percent data coverage refer to? Percent of daylight hours? Percent of geographical regions?

It refers to geographical region and this is now included in the sentence.

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15) Section 3.1. (Or perhaps there is a more appropriate section elsewhere?) Can something be said about the source of the plume, i.e., was it due to agricultural fires or agricultural fires that turned into forest fires? It would be useful to place these results into a broader context and to associate cause of the plume to the impact on the optical properties. This information may be more appropriately placed in the Introduction section of the paper.

We have included some more information based on Stohl et al (2007) in the introduction about the sources.

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16) Section 3.2.1. Define Angstrom exponent and describe how it was calculated.

We have defined the Ångström exponent. The calculation of the exponent is based on the available wavelengths for the different instruments at the different sites. This is a standard procedure. and there should not be necessary to go into details about the calculations. The available wavelengths are given i Table 1.

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17) Table 3: Is only the May 2006 AOD at ALOMAR at a wavelength of 320 nm? Each value measured at that wavelength should be indicated. Also, please provide standard deviations with the mean values to give the reader a sense of the variability of the measured AOD.

Regarding the AOD measurements at ALOMAR, only values for 320 nm is available during the pollution event as the Cimel instrument unfortunately were away for calibration in this period. Thus each value measured at 320 nm is indicated as it was only during the episode the instrument was away. The other values representing the means and typical Arctic haze are for 500 nm.

After a thorough consideration we have decided to include mean values for May instead of yearly means. We think that this is much more relevant for the episode and serves as a better reference for the high levels observed in May 2006, which was our intention. The standard deviations are included. After a revised consideration we find it misleading to use yearly means as we have done, as the data for some of the sites are incomplete with respect to time (the data are campaign data, further the time period with available data from Arctic sites vary a lot due the polar night). Thus the data we have presented are not harmonized and a comparison among the sites as well as between the episode and the reference will be somewhat misleading.

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18) Figure 4: For ease of comparison between sites, make all y-axes cover the same



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range for AOD and Angstrom exponent.

This is corrected.

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19) p. 9529, line 21: Describe in more detail what is meant by “Eˇ the AOD decreased slowly due to the stable conditions Eˇ .” Is this referring to a stable atmosphere with little deposition or vertical mixing occurring?

Yes, this specification is now included in the text.

———————

20) p. 9531, section 3.2.2. and Table 4: What is meant by “volume fraction”? Is this the fraction of the total aerosol volume that exists in a particular mode? Clarify in the text and table caption.

Yes, and this clarification is now included in the text.

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21) p. 9531, section 3.2.2. and Table 4: Is the accuracy of the retrieved parameters such that 3 and 4 significant figures are warranted? It is stated in the text that “Eˇ the retrieval of the particle volume size distribution is adequate Eˇ .” Please quantify the un-certainty of the retrieved parameters. Table 4 should include standard deviations with the mean values to indicate the variability observed.

The parameters are retrieved by non-linear curve fitting and the standard error from the fitting procedure is now included in the table. The numbers of significant figures are adjusted according to this. We will like to comment that it is our opinion that this makes the Table to detailed, but we are willing to do this if this is really wanted. See an example of the first column below.



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Site Parameter* Minsk rv1 0.160 (2.10-3) σ1 0.42 (1.10-2) Volume fraction, mode 1 0.040 (2.10-4) rv2 1.96 (3.10-2) σ2 0.49 (9.10-2) Volume fraction, mode 2 0.079 (8.10-4) rv3 7.1 (1.7.10-1) σ3 0.43 (2.10-2) Volume fraction, mode 3 0.88 (8.10-2) SSA440nm 0.92 (2.10-2) SSA1020nm 0.81 (210-2) N data 114 (63)

———————

22) Table 4 caption: Change to “Eˇ for the inversions for AOD440nm $>$ 0.5 andEˇ .”

Done

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23) p. 9531, line 28: Why are the radii for the fine mode larger near the source than at Hornsund? Is the retrieved median radii accurate to +/- 0.04 um, i.e., the difference between values measured at the three locations? The diameter would be expected to increase with time due to processing during transport.

According to the fitted parameters and their standard errors, the radii in Minsk and Toravere are significantly different from the radius in Hornsund. However it is not just the radius that is important. Also the geometric mean, and for non-linear curve fitting the parameters is not independent of each other.

It is correct that condensation will increase the radius. Further, it is the largest aerosols that are most influenced by deposition during the transport. Accordingly the total effect of the transport on the mean radius is not obvious, and one effect might be that the size distributions are shifted towards smaller radii. Furthermore, the geometric mean for Hornsund is high indicating a wider range of the radii here than in Toravere.

We will also like to point to the fact that it is important for this analysis that the instruments are equal and that the methods are the same for all sites. Which is the case for the AERONET sites. As there are no more data available for chemical or physical characterisation of the aerosols at the AERONET sites, we find it difficult to go further



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into details on this issue.

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24) p. 9532, lines 15 - 16: “Eˇ .the volume size distributionEˇ has values comparable toEˇ ” Exactly what values are being referred to here? State these explicitly.

We are referring to the parameters (radii, volume fraction, geometric mean, full characterisation of the size distribution. The sentence is rephrased to clarify this.

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25) p. 9532, lines 26 - 28: “Eˇ could explain the differences in the size distributionEˇ ” State what differences are being referred to.

We have changed the first sentences of the paragraph to make this clearer.

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26) p. 9533, line 10: SSA calculated using Mie theory and measured composition will be very sensitive to the approach used to account for the dependence of scattering on RH. Exactly how was hygroscopic growth taken into account?

We have added the following sentence: The hygroscopic growth is taken into account for sulphate, nitrate, organic carbon, and sea salt aerosols according to Myhre et al. (2007).

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27) Figure 6. It would be useful to put SSA values calculated from the Zeppelin data on this plot for comparison to the AERONET retrieved values.

This is now included. This is an average value for the days 30 April – 7 May as it is based on in situ measurements at the Zeppelin observatory as described in section 2, and this is only weekly filter samples. We have also updated the figure caption.



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28) p. 9533 - 9534, discussion of difference in SSA between source and distant regions: Two reasons are given for the higher SSA observed at the Arctic sites relative to the sites closer to the source regions: deposition and RH. Deposition is expected to (and, indeed did according to the earlier discussion) primarily affect larger supermicrometer particles for the transport times in this study. BC emitted from biomass burning is associated with smaller, newly formed particles. Is there evidence for significant BC mass in the larger size range as suggested here? RH is stated to decrease SSA, presumably by increasing light scattering but not affecting light absorption. Please provide more details on your reasoning here. Also, please expand on the statement that “The increase of SSA can also be a result of condensation and/or formation of secondary organic aerosols.” Does this refer to changes in mixing state for the BC? Please provide more details.

Regarding coarse aerosols, only a small fraction of absorbing materials is necessary to reduce SSA notably. We would not state that it is evidence for significant BC mass in the coarse mode although we think BC might be present to some extent, also in this mode. We also believe that there are other absorbing materials present like soil and mineral dust and these contribute to absorption in this mode. This is supported by the study of Formenti et al (2003) investigating forest fires during the SAFARI 2000 campaign.

Formenti, P., W. Elbert, W. Maenhaut, J. Haywood, S. Osborne, and M. O. Andreae (2003), Inorganic and carbonaceous aerosols during the Southern African Regional Science Initiative (SAFARI 2000) experiment: Chemical characteristics, physical properties, and emission data for smoke from African biomass burning, J. Geophys. Res., 108(D13), 8488, doi:10.1029/2002JD002408.

We have included the following sentences to give more details in our reasoning about the increase of SSA:



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“An increase in the water content of the aerosols or an increase of SOA, will both change the relative fraction of scattering and absorbing aerosol components towards a higher fraction of scattering components. Thus this will both result in an increase of SSA.”

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29) p. 9534, lines 27 - 28: Instead of simply stating that “the results are in good agreement”, quantify the level of agreement for the MODIS and ground-based AOD values (e.g.., provide an rˆ2 value).

Information about the correlation coefficients are now included in the text. There are differences in the time resolution of the MODIS data and the ground based data. Thus, in the calculation of the correlation coefficients an average of the ground based data closest to the time of the satellite overpasses are used.

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30) p. 9535, lines 2 - 4: Can the statement that “This is expected as the groundbased data from Minsk is for 500 nm” be supported by an analysis of the Angstrom exponent and a conversion of the data so that the wavelengths match up between the two instruments?

We want to present the original data and the Ångström exponent is given, which makes it possible to make the conversion. Our main purpose of the comparisons was to recognize the episode at the various sites and justify the use of MODIS data in the radiative forcing calculations. As shown in Table 1 the wavelengths used by the different instruments are very different (except for the instruments included in AERONET) thus a broad discussion and comparison of the conversions as well as the converted data is then necessary. We think that such an analysis might give some new information, but it will also make the paper considerable longer and we are reluctant to do it in this study as we think that this is not essential for the main object of the study.



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31) p. 9536, first paragraph: The comparison of backscatter coefficients measured as a function of altitude at Minsk could be improved by sticking to a comparison of maximum values in the boundary layer OR average values within the boundary layer.

We agree with the comment and change the text accordingly, using the average of the aerosol backscatter coefficient within the aerosol layer.

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32) p. 9536, last lines: Can values of the maximum ABC along with the height of the maximum value and the height of the aerosol layer be given for the European plume arriving later in the day on May 5th? This would provide an interesting comparison of a plume of European origin and the biomass burning plumes that are the subject of the paper.

We are a bit unsure about this comment, because from May 5th, we only have one profile from the evening (19.15–20:00) as shown in Figure 8. In case the referee is pointing towards May 6th, two profiles, from 12:00-12:45 and 19:15-20:00, are shown in the Figure 8. We have increased the line-width to make the latter profile more visible.

Stohl et al. (2007) have discussed the transport into the Arctic and the contribution of biomass burning versus emissions from fossil fuel combustion during the event. We do not expect to get additional information from an extended analysis of the lidar profiles from those day compared to what has already been described in that article.

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33) Section 4: Because there is such a lack of calculations of aerosol radiative forcing for the Arctic, this portion of the paper is perhaps the most interesting and most significant. Therefore, the description of the calculations should provide much more information than is currently given. For example: How is RH taken into account (dependence of scattering on RH)? What is meant by “AOD based on chemical



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composition Eˇ .. is scaled with AOD from MODIS.” Are these TOA forings? Diurnally averaged? Clear sky only? (This latter point is confusing because on line 19, the paper refers to “Eˇ some few cloudy regions.”) How was surface reflectance handled? Was one uniform value chosen for the model region or was it varied? This is of particular importance given the conclusion that “The climate effect of the aerosols in this region is particularly sensitive to the surface albedoEˇ .”

More details regarding the Section 4 are included in the manuscript.

We have added a sentence in section 3.2.2 (see above) and two sentences in section 4 to better explain the treatment of hygroscopic growth. The scaling to the MODIS AOD is also described in more detail. The model calculation of the surface albedo is described by the following sentences: “The surface albedo data used in the radiative transfer simulation is spectrally resolved and varies with vegetation and ground characteristics (Myhre et al., 2003a). Snow and sea ice content are taken from the ECMWF and are included in the calculations of the surface albedo.”

We also state that the radiative forcing is daily average TOA all-sky values.

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34) p. 9539, lines 9 and 10: Please provide standard deviations with the mean SSA values.

This is now included in the Table 4, and section 3.2.2 as we feel that this belongs to the presentation of the results rather than in the conclusion.

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35) p. 9539, line 21: Please quantify “Eˇwe find high agreement at all sites.”

As the correlation coefficients now are included in section 3.2.3 we do not think that



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this information is necessary to repeat in the conclusion.

Thank you!

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 9519, 2007.