**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Optimal estimation retrieval of aerosol microphysical properties from SAGE II satellite observations in the lower stratosphere" *by* D. Wurl et al.

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Final Response to Reviewers' Comments – # 2

Review of Manuscript: acp-2009-319 Optimal Estimation Retrieval of Aerosol Microphysical Properties from SAGE II Satellite Observations in the volcanically unperturbed Lower Stratosphere by D. Wurl, R. G. Grainger, A. J. McDonald, and T. Deshler

First of all the authors would like to thank the reviewers for their interest in this work and for the detailed comments that they have provided. All questions asked by the reviewers (RV) are answered below by the main author (DW) on behalf of all co-authors.

General comments:

RV: This is an interesting and well written manuscript dealing with the retrieval of microphysical properties of stratospheric background aerosols using SAGE II solar occultation observations at different visible and near-IR wavelengths. The novel aspect of this study is the use of the optimal estimation technique that allows combining measured information with a priori information on the fraction of the aerosol particle size distribution that has essentially no impact on the occultation observations. As the authors put it, using a priori information allows filling the 'blind spot' associated with the occultation measurements. This concept appears plausible and useful at first glance, but it is associated with several problems, that are – in my opinion – not adequately addressed in the paper, as outlined in the following general and specific comments.

All in all, the manuscript requires major revisions before becoming acceptable for publication. I also would like to point out, that the paper is very well written and structured, and I believe it will make an interesting and relevant publication once the limitations of the method are discussed in more detail.

DW: We have thoroughly revised the manuscript and taken great care to address all the questions and concerns raised in the report. In particular, we have reworded our description of the Optimal Estimation approach to explain more clearly how the difficulties inherent to the retrieval of background aerosol properties are addressed. A focus was also put on the a priori constraint. The limitations inherent to that particular choice are assessed and explained. A new section was written on the effect of bimodal aerosol on the monomodal retrieval results. Major changes have been implemented mainly in Section 1 (Introduction), Section 4 (Model validation), and Section 5 (Application to measured data). The details are given in the specific comments below. General comments:

RV: a) If the 'blind spot' is filled with a priori information, how useful are the retrievals then? This certainly depends on the fraction of aerosol particles that are below the

detection threshold. You write several times, that the occultation observations are insensitive to aerosol particles with radii less than about 100 nm. According to Fig. 2 and Table 3 the mean median radius of your SAGE II retrievals is about 70 nm, i.e. less that the rough sensitivity threshold of 100 nm. Looking at the histogram in Fig. 3b I estimate that less than 2 – 4 % of the retrieved median radii are actually larger than 100 nm, and I guess this implies that almost all of this information comes from the a priori, and not from the observations. This is a major issue and must be addressed. The higher order moments are certainly not as affected.

DW: The given value of 0.1 μm, which is often quoted in the literature (e.g. Heintzenberg et al., 1981), is an approximate threshold around which the sensitivity of optical measurements decreases rapidly toward smaller particles. To illustrate the low sensitivity of spectral extinction measurements to particles smaller than 0.1 μm two new tables were added to the manuscript, which list the fractional contribution of these particles to the total aerosol extinction (Table 1) and to the total particle number, surface area density and volume density (Table 2). These numbers demonstrate that for two of the three example size distributions the great majority (namely more than 80%) of all particles are smaller than 0.1 μm. The contribution of these particles to the total extinction is of the order of the SAGE II experimental uncertainty. L. 121-128 (Sect. 1, low sensitivity to particles < 0.1 μm) +Table 1 +Table 2

RV: b) Following the previous comment the choice of the a priori information becomes very critical here. You use in-situ observations at 41N latitude for the period 1991 to 1997. Several important issues arise from these assumptions that are not discussed in the manuscript at all. During this period the Pinatubo eruption occurred, which means, that the ensemble of in-situ measurements does clearly not provide an appropriate a priori for the retrieval under background conditions.

DW: A priori knowledge: Although the measurements were collected during a time period that covers the eruption and relaxation period of Mt. Pinatubo, only monomodal size distributions were selected and used as a priori data. These 264 monomodal

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aerosols size distributions have median particle radii between 0.02 and 0.2 μm and are clearly uninfluenced by Mt. Pinatubo.

We have added a new paragraph to the new manuscript to explain this issue: L. 326-341 (Sect. 4.1, a priori data) L. 330-334 (Sect. 4.1, non-volcanic monomodal a priori data)

RV: Secondly, using a priori information valid for a certain latitude is not appropriate for retrievals at all latitudes. Although the comparisons with in-situ observations in 1999 are done at the right latitude, section 4.2 suggests that all SAGE II observations in December 1999 ('19700 retrieved results') were retrieved with the same a priori. Issue a) can certainly not be solved with the present approach, but it should be mentioned and discussed in detail in the paper. The paper is still worth to be published, but it should actively address the issues associated with the basic approach.

DW: The manuscript has been considerably amended to discuss the a priori data and potential biases due to variation in altitude and latitude of aerosol properties. The associated passages in the new manuscript are indicated below.

The purpose of the a priori pdf is to add to the information contained in the measurements by describing the solution space as comprehensively as possible. As the loading of aerosol varies with height and latitude as the tropopause height changes, as well as with time (e.g. with season of the year or with the phase of the quasi-biannial oscillation, Trepte and Hitchman, 1992) the ideal a priori information would be a function of latitude, altitude, and time. However, given the paucity of aerosol measurements (other than SAGE) it seems more reasonable to use a broad a priori that captures the variation with height and latitude. Firstly, as the a priori becomes more specific (either spatially or temporally), the a priori variances and covariances would be expected to decrease. In the maximum a posteriori technique, this will tend to decrease the relative weight of the measured extinction in the aerosol retrieval and thereby increase he relative weight of the a priori mean state. And secondly, our experience of satellite re-

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trievals suggests that using spatially-varying a priori may produce spurious features in the retrieved fields (Deeter et al. 2003). Neither of these effects is desirable at present, as they both would complicate interpretation of the retrieval results.

The Wyoming in situ record (Sect. 4.1) comprises aerosols measured at different altitudes and different times of the year. It is therefore representative of a range of different temperatures and acidities. As these were, however, all measured at mid-latitudes ($41°$ N), they may not be entirely representative of all aerosols that may occur at other latitudes. A comparison with a series of in situ measurements taken at Lauder, New Zealand ($45°$ S, 1991-2001) shows that these southern mid-latitude aerosols are very similar to the Laramie ($41°$ N) time series (Deshler et al. 2003). A bias due to the a priori data being potentially unrepresentative of some aerosols that may occur at other latitudes can only be estimated when new measurements become available in the future. The results obtained with the height- and time-independent comprehensive a priori are shown to be fairly accurate even in the case of large measurement uncertainty (Sect. 4.2). See new manuscript: L. 427-450 (Sect. 4.3, a priori bias, comprehensive versus specialized a priori)

Variation with latitude: Due to the SAGE II measuring geometry, the great majority of all data measured in December were recorded at northern mid-latitudes, namely near $40°$ N. This means that the a priori data used in this study would be appropriate at least for the majority of all data presented here. In contrast, most of the September measurements were recorded at higher latitudes, namely near $60°$ N and S. If the retrieved aerosol properties in September were distinctly different from the December data, this could be an indication that the measured aerosols were not appropriately represented by the mid-latitude a priori size distributions. No great discrepancies can, however, be observed between the September and the December data. This means that the applicability of the current mid-latitude a priori for aerosols measured at other latitudes in the SAGE record cannot be disproved until new in situ measurements become available.

New text in the revised manuscript: L. 664-675 (Sect. 5.2, SAGE II: December versus

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September data, latitude bias?) L. 510-512 (Sect. 4.4, summary: potential a priori bias)

In summary, the comprehensive (as opposed to height- or time-resolved) a priori probability density functions were found to be appropriate for retrieving aerosol properties from synthetic measurements, even in the case of large extinction uncertainty and in the case of small-mode-dominant bimodal aerosols (with a few exceptions that are named in the paper). A bias due to the Wyoming data being potentially unrepresentative of aerosols at other latitudes cannot be detected in the retrieved results. At present, the mid-latitude in situ measurements provide the best prior estimate we have, and the retrieval results seem to confirm the validity of their use.

New manuscript: L. 704-710 (Sect. 6, comprehensive a priori, a priori bias)

Specific comments:

RV: Page 23720, line 26: 'For instance, by scattering a large portion . . .'. I think 'large portion' is an exaggeration. If I'm not mistaken the typical radiative forcing of stratospheric background aerosol is less than 1 W / m2, which is important, but clearly not a large portion of the incoming seasonally and globally averaged solar irradiance (342 W / m2).

DW: The objection is justified. The passage was meant to say that small sulphuric acid particles are very efficient scatterers. The passage describing the climatic impact of stratospheric aerosols has been changed to: "Stratospheric aerosols are known to play an important role in the climate system because they can influence the global chemical and radiation balance in the atmosphere in a number of ways (McCormick et al., 1995; Solomon, 1999). In the aftermath of large volcanic eruptions stratospheric aerosols have a significant impact on the Earth's radiation balance for several years after the eruption. The observation that stratospheric sulphuric acid aerosol can exert a cooling effect on tropospheric temperatures (e.g. Pueschel, 1996) has even stimulated the idea of deliberately introducing aerosols to counteract climate warming caused by anthro-

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pogenic emissions of greenhouse gases. This area of research, called geoengineering by sulphate aerosols, is receiving increasing attention (e.g. Rasch et al., 2008a,b; Tilmes et al., 2008). During volcanically quiescent periods, when stratospheric aerosol can be characterized as in a background state unperturbed by volcanism, the direct radiative impact of stratospheric aerosols tends to be rather small. However, these particles may also play a role in the nucleation of near tropopause cirrus, and thus indirectly affect radiation (Kärcher and Ström, 2003; Penner et al., 2009). Stratospheric background aerosols also play an important role in the chemical balance of the stratosphere. At mid-latitudes they affect the ozone balance indirectly by interacting with both nitrous oxides (Fahey et al., 1993) and chlorine reservoir species. For instance, NOx increases under low aerosol loading conditions and induces ozone loss from the nitrogen catalytic cycle (Crutzen, 1970). In the polar stratosphere the small aerosol particles provide condensation sites for polar stratospheric clouds which then provide the surfaces necessary to convert inactive to active chlorine leading to polar ozone loss. These examples provide an insight into the intricate interactions between stratospheric aerosols and the climate system."

L. 40-64 (Sect. 1, climatic impact of stratospheric aerosols)

RV: Page 23724, line 10: I'm surprised to read that the imaginary part of the refractive index is zero. It's quite small, OK, but I don't think it's zero. In the introduction you also write 'Through scattering and ABSORPTION of . . .'

DW: The sentence concerning the imaginary refractive was changed to read "The imaginary part of the refractive index (describing the absorption) is practically zero and hence extinction is equivalent to scattering." The introduction has been changed accordingly (as detailed above). L. 182-183 (Sect. 2, refractive index, no absorption)

RV: Page 23726, line 2: 'to particles with radii less than 100 nm'. This threshold appears several times in the paper, and I wonder how robust it is. Considering that almost all of the retrieved sizes are below this threshold (see general comment above)

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it would also be very useful to provide a more quantitative estimate of the contribution of these smaller particles to the observed extinction signal. This can be easily done using Mie calculations. RV: Page 23731, line 13 and Fig. 3: The vast majority of the retrieved median radii is smaller than 100 nm, i.e., below the sensitivity threshold. I suggest estimating what fraction of the derived sizes actually affects the observations.

DW: The calculations were done and the results described above. L. 121-128 (Sect. 1, ARP, low sensitivity to particles < 0.1 μm) +Table 1 +Table 2,

RV: Page 23732, line 15: 'they are both spherical and homogeneous'. This is a minor point, but can we really know that they're truly spheres? Such a statement cannot be verified, I believe.

DW: According to Torres et al. (1998) for sulphate aerosol models the assumption that the droplets are spherical is justified since surface tension forces liquid sulphuric acid particles resulting from the gas-to-particle transformation process to produce spherical shapes. This reference was cited in the text. L. 174-176 (Sect. 2, spherical particles)

RV: Page 23739, line 32: The SPARC report was compiled and edited by Larry Thomason and Tom Peter. I suggest they should be listed as authors (or at least editors) of the report rather than the SPARC steering group.

DW: The suggested change was implemented. L. 116 (Sect. 1, cite Thomason and Peter, 2006, for SPARC report)

RV: Page 23741, caption Table 1: 'these correlation coefficients are all significant'. Significant at what confidence level?

DW: Given the large number of measurements (namely approximately 230) these correlation coefficients are all significant at $p < 0.05$ % (Taylor, 1939, Table C). This information was amended to the table caption. L.371+ Caption to Tab. 3 (Sect. 4.2, significance level of correlation coefficients)

RV: Page 23745, Table 5, (2)/PCA: I don't understand '(15-20)+50', guess there's

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something wrong here?

DW: Table 7 provides a list of uncertainty estimates associated with aerosol properties all retrieved under non-volcanic conditions but using different retrieval techniques. The "+" indicates that a given value is an estimate of partial errors only and that the total error is expected to be higher due to other disregarded uncertainty components. The uncertainties in surface area density (A) as reported by Steele et al. (1999) and Steele and Turco (1997), for instance, account for propagated random errors only. The total errors are expected to be higher by about 50% due to disregarded systematic (method bias) errors and contributions from particles smaller than 0.1 μm (see table caption). For more clarity the table item was changed from " (15-20)+50 " to simply " (15-20)+ ".

L. 630-663 (Sect. 5.2, SAGE II: comparing uncertainties) Caption of Table 7

Typos etc.:

RV: Page 23721, line 13: I suggest rearranging words from 'retrieval aerosol problem' to 'aerosol retrieval problem'

DW: The sentence was changed to "These and other observations gathered in a recent assessment of stratospheric aerosol properties lead to the conclusion that "significant questions remain regarding the ability to characterize stratospheric aerosol during volcanically quiescent periods, particularly in the lower stratosphere" (Thomason and Peter 2006)." L. 113

RV: Page 23739, line 15: 'trueber' -> 'tr\"uber' and 'Metalloesungen' -> 'Metall\"osungen' Interactive comment on Atmos. Chem. Phys. Discuss., 9, 23719, 2009.

DW: Done. L. 839 (Literature, Mie, 1908)

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/9/C11381/2010/acpd-9-C11381-2010-

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supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 23719, 2009.