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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 – # 1

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.

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General comments RV: Even if we are convinced that this approach is very valuable, it seems to us that the application of this method has not been performed with the necessary care, and that the authors do not take into account the limitations of their choice of a priori information. At least, these aspects are not discussed at all in the paper. By neglecting the limitations and approximations inherent to the choice of a priori information, they go, to our opinion, to too fast and somewhat biased conclusions. Our argumentation to support this opinion is developed in the specific comments.

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.

Specific comments

RV: [L. 7-14, p. 23729; l. 22-26, p. 23731; l. 17-20, p. 23733; figs 7 and 8 :] The introduction of information results logically in a reduced value of the uncertainty, but it is important to keep in mind that this estimate of the uncertainty may be biased if the information used a priori is not fully relevant. The authors use as a priori the Wyoming time series which is a highly valuable source of information, but taken at a fixed latitude of 41°N. Examination of the variation in altitude and latitude of extinction show global structures following roughly the isentropics. If I understand well, this kind of variation has not been taken into account in the choice of a priori. Moreover, the authors mention that they computed the a priori information from in situ measurements taken in the time period May 1991 till October 1997, which contains the whole relaxation period of one of the most important volcanic eruption of the century ! This choice is quite surprising (and basically inadequate) while the objective is to characterize the aerosol microphysics in nonvolcanic conditions. Even if measurements bring information about aerosol microphysics, what results in a decreased uncertainty, the information content provided by measurements concerns big particles, and not very thin particles in the Rayleigh limit of scattering that they are unable to discriminate. Hence, a priori knowledge that would be not relevant for very small particles, will not be "corrected" by the information coming the extinction measurements.

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 aerosols size distributions have median particle radii between 0.02 and 0.2 μm and are clearly uninfluenced by Mt. Pinatubo. From these measurements a general comprehensive a priori was generated which captures the variation with altitude and time.

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: This problem may induce a bias in the retrieved quantities, especially at geolocations for which the aerosol population found at Wyoming would be less representative or at altitudes very different from this of the first iteration (See l. 14, p. 23729). Actually, the illustration of the contribution of the a priori in Fig. 7 and 8 clearly shows that this contribution is not representative for very different altitudes. A detailed discussion about these aspects should be at least included in the paper. The last paragraph in section 4 should also be qualified by mentioning that the a priori information describing the thin particle contribution might be not optimal for the considered geolocation and time. As a conclusion for this point, adding information content leads to a reduced value of the uncertainty, but a reduced value of the uncertainty only corresponds to a better precision on the aerosol microphysical properties if the a priori information reflects the reality in a correct way. My opinion is that the authors could improve their use of in

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situ data to better match the reality. They are of course limited by the very restricted number of available in situ data sets, but they should at least discuss these limitations.

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 retrievals 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 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.

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

RV: Further, they should be very cautious while comparing uncertainties provided by other approaches in section 5.

DW: The authors agree with the reviewers that care has to be taken when comparing uncertainty estimates associated with results that were obtained through different retrieval approaches. Such a comparison cannot give conclusive evidence of the accuracy of the achieved results, because realistic uncertainty estimates are in practice often difficult to obtain. As under certain circumstances not all biases can be reliably estimated, some uncertainty estimates may represent partial errors only. Nonetheless, the uncertainty estimates can give an indication of the quality of the retrieved results provided that the scope of application (included aspects and expected additional uncertainties) is taken into account. Given that the OE retrieved uncertainty estimates tend to be a fairly realistic estimate of the true errors, such a comparison with other uncertainty estimates seems interesting.

New paragraph in the (new) manuscript: L. 630-663 (Sect. 5.2, SAGE II: comparing uncertainties) L. 711-725 (Sect. 6, retrieved uncertainty estimates)

RV: [L. 19, p. 23734 till l. 1, p. 23735] The authors present some intercomparison with PCA results and with data set derived by Bingen et al., and conclude that their estimates are closer to the correlative in situ measurements. Do I understand well that they use as in situ reference data the Wyoming time series already used for computing the a priori? If this is the case, it is clear that the OE results are likely to be closer to the in situ data set, and that this comparison is not suitable to validate OE results.

DW: The in situ reference data were measured at the same latitude as the a priori data, but in 1999 (see original manuscript, caption to Fig. (7) and explanation in the text on page 23734, line 22) and hence they are different from those measurements that were used to generate the a priori pdfs (1991-1997 data).

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RV: Even if the time series has been split up in two sets of profiles, one for the computation of the a priori, and the other one as reference data set for comparisons, the comparison is biased because all these profiles concern the same location, and they do not take into account altitude/latitude dependence of the extinction profile. If the authors use other sources of in situ measurements, it should be clearly mentioned.

DW: (These concerns have been addressed above.)

RV: L. 4-5, p.23724 : It should be mentioned that extinction is calculated using Mie's theory because aerosol particles are assumed to be spherical.

DW: We have changed the sentence from "It can be calculated using Mie's theory of light extinction (Mie, 1908), where extinction is the sum of scattering and absorption." to "As tiny sulphuric acid particles can be assumed to be spherical (Torres et al., 1998) and homogeneous, the extinction coefficient can be calculated using Mie's theory of light extinction (Mie, 1908)." L. 174-176 (Sect. 2, spherical particles)

RV: L. 5, p.23726 : Actually, the complexity of the aerosol retrieval problem mainly arises from the ill-posedness of the inversion problem of retrieving N, R, S from Eqs. (1,6), and from the theoretical limitation related to Rayleigh scattering: Extinction is independent of the size particle in the Rayleigh limit of scattering, hence size information of very small particles with respect to the wavelength, cannot be retrieved from optical measurements.

DW: We have revised the description of the aerosol retrieval problem and amended the manuscript to include the above points.

The relevant passages in the new text are: L. 1-9 (Abstract: Aerosol Retrieval Problem, ARP) L. 119-135 (Sect. 1, origin of "aerosol retrieval problem") L. 231-235 (Sect. 3, ill-posed problem) L. 679-682 (Sect. 6, aerosol retrieval problem)

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

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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).

L. 121-128 (Sect. 1, low sensitivity to particles < 0.1 μm) +Table 1 +Table 2

RV: L. 23-27, P. 23730 : There is a much more fundamental reason why A, V, Reff are expected to be better retrieved than N, R, S. The reason is that A, V and Reff are integrated quantities, whereas N, R and S are functions in the integral (6) with N(r) given by (1), to be retrieved by inversion of the extinction. Fluctuations and uncertainties on N, R, S are smoothed out during integration, what explains the higher stability of A, V and Reff. Conversely, small fluctuations and uncertainties of the extinction, which is an integrated quantity, give rise to a highly amplified fluctuation of the functions in the integral, i.e. on N, R and S.

DW: We have amended the manuscript to include this point. L. 374-382 (Sect. 4.2 integrated properties less sensitive)

RV: [§4.1, figures 1, 2,:] : Are the test beds are really representative of the non volcanic situation that the authors intend to simulate? The authors mention that they consider 264 monomodal aerosol size distributions originating from in situ measurements by Deshler. What are these measurements ? Are they size distributions at fixed points (at which height ?) or profiles ? At which period ? Do they possibly consider only the thin mode of bimodal size distributions ? In the Wyoming time series, it can be observed that monomodal distributions are mainly used from the early seventies until the time period 1990-1995, and bimodal size distributions are used in the period 1980-1985 and after 1990, period including the non volcanic periods studies in the present paper. Hence, it seems that, either the authors don't use size distributions related to the studied period, or they possibly extract thin modes from bimodal distributions provided by the time series. In the first hypothesis, it is not clear if the used profiles concern a non volcanic period : the number of monomodal size distribution found after 1997 is much less that 264. Concerning the second hypothesis, see next remark. Can the authors give more information about his point ?

DW: See above comments for an explanation of the a priori data. Although they were measured during the Pinatubo period, the selected distributions are monomodal and clearly uninfluenced by the volcanic eruption. We have added a new paragraph to the manuscript to clarify these points: L. 326-341 (Sect. 4.1, a priori data) L. 330-334 (Sect. 4.1, non-volcanic monomodal a priori data);

RV: [L. 9-16, p. 23733 :] I am not convinced by the argumentation of the authors concerning the bimodal error : a careful study of the Wyoming time series also shows that, in the case of an aerosol population characterized by a bimodal size distribution (thin +coarse ones), the typical particle number densities may differ from several orders of magnitude: the number density is much larger for the thin mode than for the coarse mode. However, a calculation of the partial extinction corresponding to each mode shows that the respective contributions of both modes, although the very different ranges in number density values, may be on the same order of magnitude. In Bingen et al., Ann. Geophys., 2003, the authors consider a retrieval technique using a lognormal distribution that favours the coarse mode, well discerned by optical measurements at the SAGE II wavelengths. The authors discuss the comparison of their results using partial number densities and illustrate the ability of their retrieval technique to describe the “coarse part” of the size distribution, and its limitations concerning the description of the thin particle contribution. Baumann et al., JGR, 2004, consider another approach where they compute a correction for thin particles. Both approaches result in very different values of the aerosol parameters, e.g. significantly higher values of the median radius. The authors of the present paper should revise their discussion about bimodal error using these papers that illustrate, together with the complementary approach of their own paper, how both monomodal coarse and thin modes may contribute equally to the extinction and how reducing the size distribution to a monomodal description may affect de retrieval. This also emphasizes again the need to a reliable estimate of the a priori for the thin mode.

DW: We have amended the manuscript to include a detailed analysis of how small-mode-dominant bimodal background aerosols influence the aerosol properties that are retrieved using a monomodal forward model. L. 451-506 (Sect. 4.4, bimodal aerosols, Forward Model (FM) bias)

The results were compared to those by Bauman et al., JGR, 2003 who did a similar analysis. L. 498-506 (Sect. 4.4, FM bias, comparison with Bauman et al., 2003)

Other associated passages: L. 22-29 (Abstract: bimodal aerosol, Forward Model (FM) bias), L. 699-703 (Sect. 6, FM bias, bimodal aerosol)

RV: The author should also reexamine the adequation of the terms “medium sized aerosol” for the scenario given and/or the choice of their simulation scenario at l. 3, p.23733 based on this discussion.

DW: Medium sized aerosol was meant in respect to the background range. We have changed the description to “typical background aerosol”. L. 421 (Sect. 4.3, Error analysis, “typical background aerosol”)

RV: [L. 26-27, p. 23734 :] I am surprised that the deviation increases while the altitude decreases: at lower altitude, the median and effective particle radii increases, so that the a priori information taking into account thin particles added to OE should bring less information content. Hence, I would expect that results provided by both methods tend to converge at lower altitude. Can the authors comment on that ?

DW: A comparison with the associated in situ number densities and monomodal median radii helps to understand the observed differences: the particle radii decrease from 0.06 μm, at 19 km to about 0.02 μm, at 13 km. Simultaneously the number density increase strongly from about 10 to over 100 particles per cm3 at 13 km (near the tropopause). Decreasing sizes mean less information content and hence the different method results can be expected to differ more. Convergence between the method results can be observed where the particles are largest, namely near 24 km where the

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median particle radius is about 0.08 μm.

L. 595-602 (Sect. 5.2, OE/PCA/in situ: disagreement where small particles) L. 613-614 (Sect. 5.2, OE/PCA/in situ: convergence where large particles)

RV: [Section 5 :] As mentioned above, the authors should also compare their results with Baumann's results (See twin paper by Baumann et al., J. Geophys. Res., 2003). As already written above, these authors study the bias due to assumption of unimodality by considering a wide set of in situ data, and propose bias correction coefficients derived from the in situ data sets.

DW: (These concerns have been addressed above.)

Technical corrections:

RV: [Section 1 :] For the completion, the authors that intent to study stratospheric aerosols in non volcanic conditions should present a comprehensive overview of the literature published on this topic and should mention works about the presence of meteoritic contributions (for instance, Murphy et al., J. Geophys. Res, 2007) and of soot (for instance, Renard et al., J. Geophys. Res., 2008).

DW: We have rewritten and considerably expanded the introduction (Sect. 1) to present a comprehensive overview of all important aspects related to the study of stratospheric aerosols in the volcanically unperturbed lower stratosphere. L. 40-149 (Sect. 1, Introduction)

RV: [§4.2, first paragraph :] The algorithm is applied to SAGE II measurements recorded in December 1999. What are the 19700 retrieved results mentioned in the text ? How do they find 19700 size distributions in this month ?

DW: The approximately 19700 events are the number of spectral extinction measurements recorded at different altitudes during 31 days with 30 sunrise/sunset events per day (relative to the spacecraft). To give an example: 31 (days) x 2x 15 (sunrises+sunsets) x 20 (altitudes) = 18600 This is how we get this large number of retrieval results. Associated passages in the text: L. 194-199 (Sect. 2, SAGE II measurements in general) L. 516-518 (Sect. 5.1, SAGE II, 19700 sets of spectral extinction data),

RV: [L. 14-16, p.23732 :] This strong statement should be qualified ! The spherical approximation is probably very good, but is still an approximation. And the Mie approximation is still an approximation, hence the Mie solution cannot be exact !

DW: The formulation has been changed to: Since the tiny sulphuric acid and water droplets of background aerosols found at temperatures above the frost point are expected to be spherical and homogeneous, deviations from Mie theory are assumed to be small. L. 405-408 (Sect. 4.3 error analysis, deviations from Mie theory)

RV: [L. 22, p. 23734, figures 7 and 8, frames b/d/f/h] The authors should specify how they compute the relative difference between PCA and in situ: (in situ -PCA)/ in situ ? (in situ – PCA)/PCA ? Something else ?

DW: We have amended the manuscript to specify how the relative differences were calculated, namely (PCA-insitu)/insitu. (Previously, it was calculated as (insitu-PCA)/insitu, but we have changed this to present the comparison in a more intuitively obvious way.) L. 589-591 (Sect. 5.2, Eq. 16)

Similarly, the differences between the retrieved OE results and the PCA results in Fig. 6 (original manuscript) or Fig. 13 (revised manuscript) were calculated as (PCA-OE)/OE. L. 564 (Sect. 5.2, Eq. 15)

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/9/C11369/2010/acpd-9-C11369-2010-supplement.pdf

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