

General comments :

For sure, this is a very interesting approach bringing potentially significant improvements in the characterization of aerosol size distribution. Nevertheless, the problem of optical inversion of extinction measurements to size properties remains, a very delicate issue that has to be treated very cautiously.

In this paper, the authors intend to improve the current estimates of aerosol microphysical properties (size distribution, surface area and volume density, and effective radius) under non-volcanic conditions. In such a case, the aerosol population is expected to be mainly composed of thin aerosol particles. The authors use the SAGE II data measured in 1999 as extinction data set. At the SAGE II wavelengths, scattering by very thin particles (radius smaller than about 0.1 micron) mostly occurs in the Rayleigh regime, for which the size discrimination is almost impossible. In order to alleviate the ill-posedness of this problem, the authors add a priori information about the thin aerosol mode. This results in a reduced uncertainty and should give a better insight in the microphysical properties of the aerosol population. As a priori information, the authors use available in situ measurements (that provide a good size resolution for thin particles), namely the time series measured at Wyoming, Laramie (41°N).

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.

Specific comments

[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 non-volcanic 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. 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 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. Further, they should be very cautious while comparing uncertainties provided by other approaches in section 5.

[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. 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.

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.

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.

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.

[§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 than 264. Concerning the second hypothesis, see next remark. Can the authors give more information about his point ?

[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. 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.

[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 ?

[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.

Technical corrections:

[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).

[§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 ?

[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 !

[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: $(\text{in situ} - \text{PCA}) / \text{in situ}$? $(\text{in situ} - \text{PCA}) / \text{PCA}$? Something else ?