

Interactive comment on “Statistical estimation of stratospheric particle size distribution by combining optical modelling and lidar scattering measurements” by J. Jumelet et al.

J. Jumelet et al.

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#This paper presents an interesting new method for retrieval of size parameters of stratospheric aerosol and polar stratospheric clouds (PSC), based on lidar measurements at 3 wavelengths. Since many lidar stations are capable of performing such measurements, the developed retrieval method could have a broad potential usage and results, in terms of a better characterisation of the observed particles, will certainly have scientific relevance. The paper is well written and the methods described in an easily understandable way with an appropriate abstract and clear conclusions, good illustrations. Although I do have a few suggestions for improvements to the analysis and discussion, as explained below, I would recommend the paper to be published in ACP after these comments have been taken into consideration.

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Comments:

Page 8921, first paragraph and in general to the retrieval method: the authors seem to ignore that the STS equilibrium composition mentioned nowhere in the paper. Instead, the assumed H₂SO₄ mixing ratio is included implicitly through the total number density of is calculated, based on the assumed condensed H₂SO₄ mixing ratio - this is particles (N₀) - a parameter in the size distributions which is obviously very difficult to constrain from the measurements, and N₀ is therefore allowed to vary within at least two orders of magnitude in parameter space. However, N₀ is a rather stable parameter with values around 10 cm⁻³ as can be seen in Figure 3, panel b in Deshler et al. (2000). I could imagine that you would obtain a more stable retrieval method if you assume a specific value of the condensed H₂SO₄ mixing ratio (as you do it for gaseous HNO₃ and H₂O) and use this value (together with the variable temperature, pressure, and partial pressures of HNO₃ and H₂O) to calculate the variable STS composition and thereby the variable real part of the refractive index. I would assume that you can get a good handle on the background sulphate aerosol loading, e.g. from your own lidar measurements under warmer conditions when the background stratospheric aerosol is made up of binary H₂SO₄/H₂O particles. Simple methods such as presented e.g. by Pinnick et al. (JGR 85, 4059-4066, 1980) could be used to derive good estimates of the sulphate mass content (or I guess you could use your own retrieval method, applied to background stratospheric aerosol measurements). Nearly constant H₂SO₄ mixing ratio values around 1 ppbv (at least for the 1996-observations, perhaps a little lower for the 2005-observations) would not surprise me, but certainly not something that varies by two orders of magnitude as does your N₀ values in the parameter space.#

>> This comment is correct. The reviewer is proposing to simplify the algorithm based on some a priori knowledge of the H₂SO₄ mixing ratio. However, since the algorithm is able to correctly retrieve the three size distribution parameters, there is no need to do so : specify N₀ indirectly and retrieve only two parameters. The fact that N₀ is searched over two orders of magnitude is not a problem for the algorithm, because

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the retrieved values are always found around 10 cm⁻³ in the cases considered here. The retrieved aerosol volume densities and H₂SO₄ mixing ratios are fully consistent with the observations. Finally, we want to keep the algorithm as general as possible, because there are optically dominant PSC modes with N₀ even smaller than 1 cm⁻³. It is true that there was no information provided on the implicit dependence of H₂SO₄ on the specified size distribution. The text has been modified: The model is initialized with a value of condensed H₂SO₄ mixing ratio, the total (gaseous + condensed) amounts of HNO₃ and H₂O and it then redistributes HNO₃ and H₂O between the gas and the condensed phases according to the calculated particle composition. The iterative procedure of the equilibrium composition calculation ensures that the gas phase and condensed HNO₃ and H₂O is equal to the initial total HNO₃ and H₂O. The model then derives the condensed aerosol mass concentration (or aerosol volume concentration) which is compared to the value of the specified particle size distribution. The H₂SO₄ mixing ratio is adjusted iteratively (and so is the composition) in order for the calculated aerosol volume density to match the one of the input size distribution.

#Once the N₀ value does not enter the calculations of the STS composition and the refractive index, the CR calculations become independent of N₀ as it, of course, should be assuming a lognormal distribution. Furthermore, the five optical quantities (3 BC + 2 CR, top on page 8922) are not independent. Since you can derive the refractive index independent of N₀, I must say that I find it more straight forward to apply the techniques as described on page 8922, line 2- 6 and section 5, page 8933, line 23-24, i.e. using only the two CR to derive the median radius and geometrical standard deviation, of course applying your cluster filtering and statistical error analysis, and then derive N₀ from the BC’s. Anyway, it would be nice if you also applied this technique to the OPC data and show a figure like Fig 5 for this case. I think you could deepen the discussion/conclusions on the refractive index. To me the most likely explanation for the mismatch between measured and retrieved N₀ values lies in too low m-values. It would be interesting to see if you would do an analysis as described above on the OPC data and you would reach the same conclusion that the retrieval is improved by

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increasing the values of the refractive indices (see also a discussion in Larsen et al., JGR 105, 1491-1502, 2000 on this issue).#

>> No (and also r_m and σ) enters the STS composition. Therefore, we cannot a priori simplify the calculation even if in most cases, this simplification can be done. The Larsen et al. reference has been added. It is true that backscatter coefficients and color ratios are not independent sources of information. Nonetheless, all our tests showed that the use of color ratios on the top of the 3 BCs improves the quality of the retrieval. The mathematical dependency of N_o in the expression of the backscatter coefficient is such that, when forming the Color Ratios, the median radius and standard deviation parameters are given more weight in the retrieval process, because the influence of N_o in CR only appears in the refractive index. This has to be connected to the low N_o sensitivity to the backscatter (as compared to r_m and σ), making it the most difficult parameter to retrieve.

Minor technical comments: Page 8910, line 9: change mode radius to median radius.
> done Page 8927, line 3: Change Hanson to Hansen (also in the reference list). >
done Figure 2: perhaps a coloured surface plot (something like Fig. 5) would be more illustrative. > We need a 3D plot to display clearly the oscillations.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8913, 2008.

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