

Dear Robert Damadeo,
we thank for your very valuable comments. We revised our paper in light of your comments (in black). The answers are shown below in red.

One important note:

If you agree we will include your comment on the etalon effect into the manuscript, see below.

Best wishes,
Pohl et al.

The authors describe a new aerosol retrieval from SCIAMACHY as well as new retrievals of various aerosol PSD parameters and compare them with other ground- and space-based measurements and retrieved parameters. Knowledge of both the amount and size distribution of aerosols is of key importance not only for climate modeling but also the retrievals of both aerosols and trace gases from many different instruments. PSD parameters are particularly important for many satellite retrievals as most instruments rely upon assumptions, rather than measurements, of these parameters for their retrieval algorithms. This paper is well organized and presented and I would recommend it for publication. The following comments are minor and only offer up suggestions for improvement or clarifications.

Pg 07, Ln 197: “For the public, we also calculate the aerosol extinction coefficient at 525 and 1020 nm to enable a comparison with other satellite aerosol products”

How is this done exactly? Is this done using the measurements at 750 nm and 1020 nm to compute the Angstrom exponent to then relate 525 nm to one of those channels?

It is calculated by Mie theory. We refer to the corresponding equation (6).

Pg 07, Ln 209: “0% relative humidity”

Does this assumption impact the data quality at the bottom of the profiles?

Yes, please see next comment.

Pg 07, Ln 210: “They are specified as a mixture of 75% sulphuric acid and 25% water.”

How does this assumption impact the results seeing as how recent measured estimates of this parameter from ACE-FTS show variability in this concentration?

To address this comment, we have added the following text:

„Both, the aerosol composition and the relative humidity, are idealistic assumptions. The percentage of sulphuric acid can vary slightly in reality (Turco et al., 1982, Steele et al., 2003, Doeringer et al., 2012). The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) even occasionally detected sulphuric acid levels of less than 50 % after the Raikoke eruption 2019 (Boone et al., 2022). The stratospheric relative humidity is usually between 0 and 10 % (Steele et al., 1981). However, we stick to these conventions because the OPAC database does not offer more realistic compositions. The resulting retrieval uncertainty was estimated by comparing retrieved PSD parameters assuming a relative humidity of 0 % and 80 %. The latter value is exceedingly high, but allows a maximum uncertainty estimate of below 15 % for the mode radius and below 10 % for the geometric standard deviation (not shown). These values can also be regarded as an uncertainty estimate due to an incorrect aerosol composition. By increasing the relative humidity, the particles absorb water vapour, which reduces the percentage of sulphuric acid. As a result, the aerosol refractive index (Palmer and Williams, 1975) changes with a similar amplitude to that of an increase in relative humidity (Hess et al., 1998).“

Pg 08, Ln 216: If N remains fixed, how is the value determined? Is a single value retrieved

somewhere else for each measurement or is a single value used for all retrievals?

The N profile is not retrieved, but is preset and never changes. To make this statement clearer, we reordered Sect. 4. The general description of the atmosphere is at the beginning („a number density profile based on the ECSTRA model“) followed by the retrieval description. Respective sentence is now:

„While r_g and s_g are derived, N remains unchanged at the initial profile for two reasons.“

Pg 08, Ln 240: “The noise covariance matrix is assumed to be diagonal, i. e., the noise is spectrally and spatially uncorrelated.”

If there is known stray light, should this be the case? Or is the stray light sufficiently small as to ignore it completely? I am guessing there is some transition region near the upper end of the retrieval range.

To address this comment, we have added the following text:

„In the absence of better knowledge, the noise covariance matrix is assumed to be diagonal, i.e., the noise is spectrally and spatially uncorrelated. Since the influence of stray light below 35 km is small, this assumption should not have a negative impact on the retrieval. The diagonal elements contain...“

Pg 16, Ln 430: “Therefore, the SAGE II and SAGE III extinction coefficients are converted to 750 nm via the Ångstrom exponent”

If you use the ~750 nm SAGE III channel for the TWE method of computing Reff, why not also use the extinction data from that channel instead of converting from the other two?

We have switched from converted extinction coefficients at 750 nm to extinction coefficients at 755 nm.

Pg 18, Ln 471: “Discrepancies are slightly higher in the tropics at altitudes below 22 km due to cloud effects”

Why not apply some rudimentary cloud filtering (or omit all data below just above [e.g., 1 km] the tropopause)?

Cloud identification is complicated. Cloud filters with stronger efficacy than applied in the manuscript could be used but at the cost of missing some aerosol-rich plumes. We therefore argue that sticking with the simple cloud filter is preferable but also mention the possible cloud contamination:

„The data with extinction coefficients greater than 0.1 km^{-1} are excluded from the comparison to reduce cloud effects. Note that this cloud filter is too simplistic to successfully eliminate all cloud contaminations. However, it prevents aerosol enriched retrievals from being incorrectly identified as clouds and excluded from the data set.“

Pg 20, Ln 500: “However, SCIAMACHY v2.0 and the SAGE II DWE approach rely on different assumptions ...”

What about the SAGE II NASA approach? It appeared that the SCIAMACHY v2.0 Reff matched those better than the DWE approach.

The SAGE II NASA approach used here (v7.0) does not provide any PSD parameters.

Pg 20, Ln 509: “... the differences in r_g and σ_g correlate with the differences between SCIAMACHY-assumed and SAGE III-retrieved number densities ...”

They correlate, but do their magnitudes align with the sensitivity tests shown in Fig. 2?

Yes, we have added the following sentences:

„If the assumed N from SCIAMACHY is greater than the derived N from SAGE III, r_g from SCIAMACHY is usually smaller and σ_g is usually larger than those from SAGE III. The magnitudes of differences align with the relative errors shown in Figs. 2 and 5.“

Pg 23, Ln 532: “Note that effective radii from SAGE III increase slightly but significantly over time. It is due an increasing median radius with a simultaneously decreasing geometric standard deviation. Such an evolution of the aerosol particle size is not observed in SCIAMACHY and both SAGE II (v7.0 NASA, DWE) data sets. This might be because in those three retrieval algorithms one of the PSD parameters is assumed to be constant.”

Is there another referenceable source that definitively shows mean radius/geometric SD systematically increasing/decreasing over this time period to show that the SAGE III data is correct and the SAGE II / SCIAMACHY data is incorrect?

Unfortunately not. The key message of this plot (new Figure number 11) should rather show differences than indicate which data set might be correct. The latter is not possible due to the lack of available data sets. We rephrased this passage based on another referee comment, who suspects these „are symptoms as opposed to the root cause.“:

„A slight but significant upward trend in the effective radius from SAGE III can be observed especially at the altitude of 21.7 km. This comes along with an increasing median radius and a decreasing geometric standard deviation (not shown). Such a significant evolution of the aerosol particle size is not observed in SCIAMACHY and both SAGE II (v7.0 NASA, DWE) data sets. A possible reason might be that in all of the latter three retrieval algorithms one of the PSD parameters is assumed to be constant, namely N_{ECSTRA} in the SCIAMACHY retrieval, the total N of 20 l/cm^3 in the v7.0 NASA retrieval, and $s_g=1.5$ in the DWE approach.“

Pg 26, Ln 615: “Thomason et al. (2010) have reported on an impact of the etalon effect on the water vapor retrieval. An additional influence of this effect on the extinction coefficient retrieval cannot be excluded.”

It is unlikely that the etalon impacts either the 520 or the 1020 nm channels in a meaningful way. An etalon is a spectral interference pattern that can change with the thickness (correlated to temperature) of the attenuator. This interference pattern will be most influential when attempting to resolve fine spectral absorption features such as with the water vapor or oxygen A-band retrievals, particularly because the temperature of the attenuator will change during an occultation. The measurement of aerosol through the 520 and 1020 nm channels does not depend on resolving any spectral features and is effectively broadband thus likely averaging out any interference patterns.

We thank you for this valuable comment. We renamed and rewrote this section and – if you agree – included your comment in the manuscript. We have additionally included the 449, 756, and 1544 nm channels.

„8.2.3 Low-distorted extinction coefficients

We compared the 520 to 1020-nm extinction ratios of SAGE II with respective 520 to 1021-nm extinction ratios of SAGE III. The latter were found to be greater due to lower $\text{Ext}(1021 \text{ nm})$ values. It is not obvious, whether the SAGE II or the SAGE III extinction coefficients are closer to the truth.

A simple explanation would be a slight overestimation of the SAGE II $\text{Ext}(1020 \text{ nm})$ values which leads to uncertainties in the effective radius. On the other hand, SAGE III measured transmissions are associated with small uncertainties due to an etalon effect, caused by a solar attenuator plate in the entrance optics. The solar attenuator was a neutral density filter where one side should be wedged by less than 1 arcmin. Due to the actual plane-parallel alignment of the filter sides, the

attenuator acted like an etalon and caused interference patterns on the charge-coupled device (CCD) image sensor.

Thomason et al. (2010) have reported on an impact of the etalon effect on the water vapor retrieval from SAGE III-M3M. The etalon induced interference pattern was most influential when attempting to resolve fine spectral absorption features such as the water vapor or oxygen A-band retrievals, particularly because the temperature of the attenuator changed during an occultation event. The measurement of aerosol from the 449 to the 1544 nm channels used for the effective radius retrieval does not depend on resolving any spectral features. It is effectively broadband thus likely averaging out any interference patterns (Robert Damadeo, personal communication).

Considering this, we cannot provide explicit reasons for the differences in the extinction coefficients of SAGE II and SAGE III, but we can emphasize that they may contribute to the offsets between the different effective radius products.“

Pg 26, Ln 628: “The retrieved median radii and geometric standard deviations should therefore be considered with caution in areas with high aerosol loading.”

This is unfortunate as these scenarios tend to be of greater interest to the scientific community. Is there any way to iterate the retrieval and update the assumptions based on other retrieved parameters such as extinction and/or effective radius?

We thank you for this good question. Theoretically, it is possible to use an independent data set as an a priori data set for the 2-parameter retrieval. In practice, there is currently no approach of how the independent data set can be used to create the a priori data set. For example, one could assume that the a priori number density must be greater in areas with enhanced extinction coefficients, e.g., after volcanic eruptions. However, there is no information on how much the number density is to be increased. Even if such an approach can be found, there are other challenges within the retrieval, e.g., the strength of the regularization depends on the particle size or the dependence of the retrieval result on the a priori result. Therefore, we have added the following text passage:

„Retrieving more accurate aerosol characteristics in areas of strong aerosol burden is of great interest to the scientific community. It requires an optimization of the a priori information. Independently observed or simulated aerosol data sets could be used to adapt the a priori aerosol profiles in the retrieval. This supposedly simple approach is challenging for several reasons, some of which are explained below.

First, there is currently no practical approach that describes how an independent data set can be used to adapt the a priori data set. Second, in layers with strong aerosol perturbations, the strength of regularization correlates in particular with the aerosol particle size. Some retrievals therefore require smaller covariance values to keep them stable. How to adapt the a priori covariance depending on the aerosol load is unknown. And thirdly, following from the previous point, the retrieval result may depend significantly on the a priori value if the a priori covariance is chosen too small.“