Consistency of Stratospheric Aerosol Measurements and Modeling: Results from ASAP and Beyond

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Introduction

The Assessment of Stratospheric Aerosol Properties (ASAP) was a SPARC-sponsored effort to present a systematic analysis of the state of knowledge of stratospheric aerosols including their precursors. It included an examination of precursor concentrations and trends, measurements of stratospheric aerosol properties, trends in those properties, and modeling of aerosol formation, transport, and distribution in both background and volcanic conditions.

ASAP found that space-based and in situ measurements of aerosol parameters tend to be consistent following significant volcanic events like El Chichón and Pinatubo. However, during periods of very low aerosol loading, this consistency breaks down and significant differences exist between systems for key parameters including aerosol surface area density and extinction. At least part of this problem arises from the fact that many parameters required for scientific or intercomparison purposes are often derived indirectly from the base measurements. This is true for space-based measurements where only bulk extinction is measured but also true in degree for most ground-based and in situ systems. Disagreements between the various data sets and models indicate that significant questions remain regarding the ability to characterize stratospheric aerosol during volcanically quiescent periods, particularly in the lower stratosphere. As reported by ASAP, models that include aerosol microphysics including formation and loss processes may provide the insight required to understand the measurement ensemble.

While ASAP highlighted the differences among the various data sets the ultimate sources of these differences were not determined. As a follow up activity, we have initiated an in-depth review of the internal and mutual consistency of three long term data sets: the Stratospheric Aerosol and Gas Experiment (SAGE II), the Halogen Occultation Experiment (HALOE), and the University of Wyoming Optical Particle Counter (OPC) including comparison to model results. Early results from this study are shown in this poster.

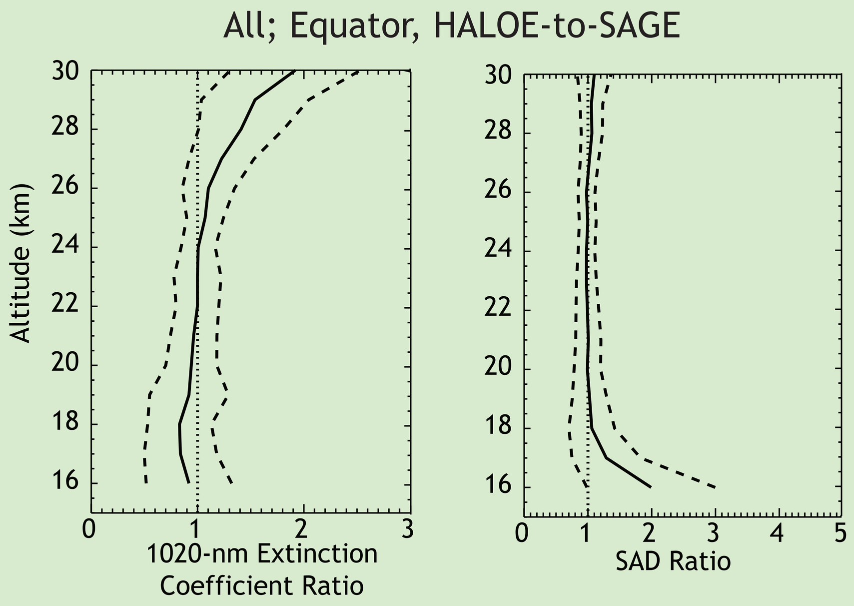
Nominal 'ASAP' Aerosol Measurement Comparisons

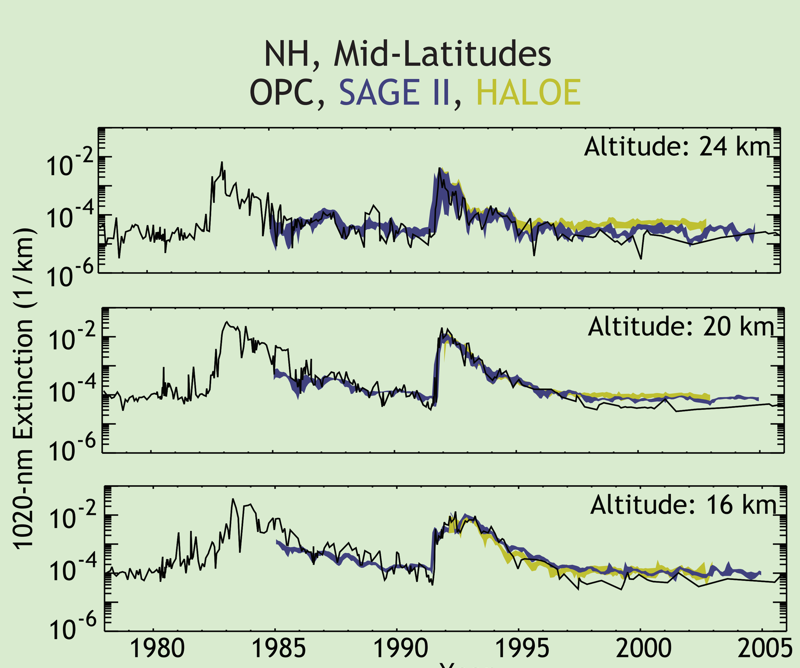
ASAP compared the long-term data sets from SAGE II, HALOE, and the University of Wyoming Optical Particle Counter (OPC) by converting the data sets to common bulk properties primarily 1020-nm aerosol extinction (measured by SAGE II) and aerosol surface area density (SAD) which is a derived quantity for all 3 instruments. The figures below (left) show time series of these parameters for mid-latitudes adapted from those shown in the ASAP report. The OPC is a solid black line while SAGE II (blue) and HALOE show the time-dependent 1-sigma range for the parameters. For HALOE and SAGE II, we use zonal mean averages (with zonal standard deviations) to increase what could otherwise be a rather sparse coincidence set. Away from time periods following volcanic eruptions, aerosol is reasonably zonal and this approach is acceptable. In addition, we show profiles of mean profiles (solid) of the extinction and SAD relative to SAGE II (below right) along with 1-sigma standard deviations (dashed) for the tropics (top) and mid-latitudes for the entire period (middle) and for only observations after 1996 (bottom).

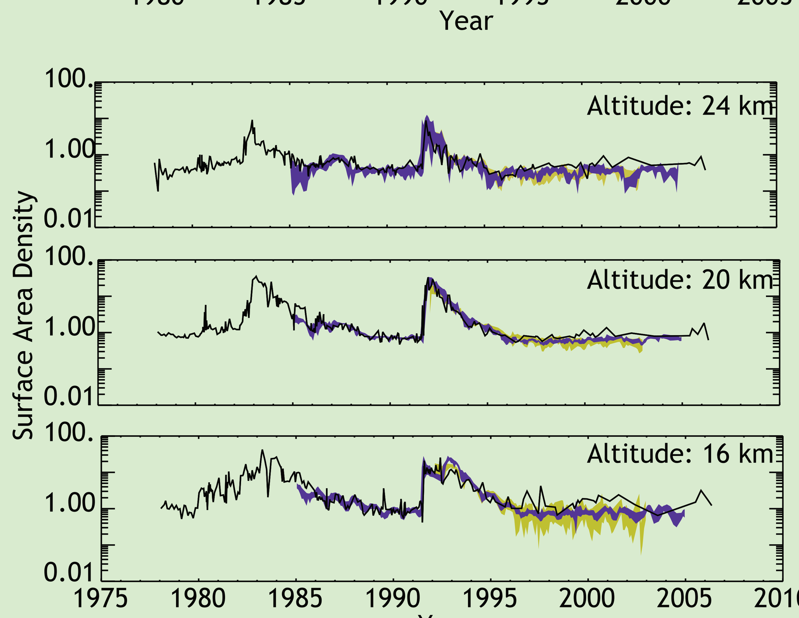
We find:

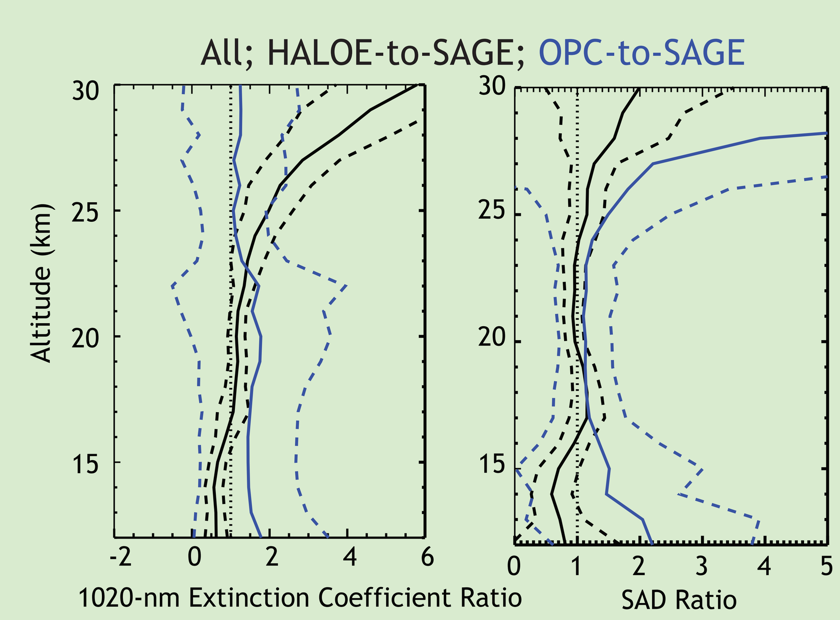
1. All quantities agree reasonably well when aerosol loading is high particularly in the post Pinatubo period (1991-1996)
2. During low aerosol loading periods (particularly after 1996), ASAP reported substantial differences in derived quantities that reach as large as a factor of 2 and, for the OPC and SAGE II, opposite in sign between extinction and SAD.
3. The OPC/SAGE II ratio at low aerosol loading are different after Pinatubo than they were beforehand even at similar extinction and SAD levels.

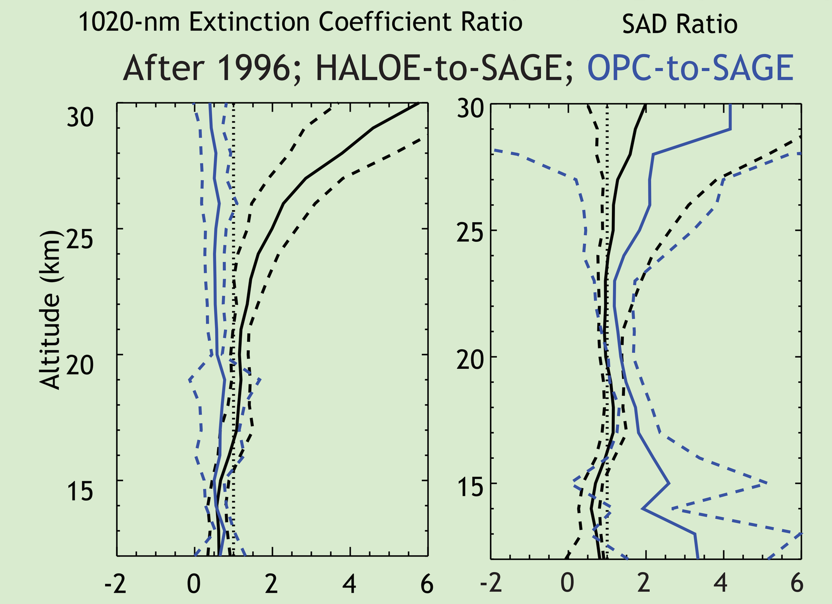
Thomason, L. W., and T. Peter (eds.)., SPARC Assessment of Stratospheric Aerosol Properties, WCRP-No. 124, WMO/TD- NO 1295, Sparc Report No. 4, 2006.











SAGE II Aerosol Measurements

SAGE II aerosol extinction measurements are well understood and validated (e.g., recent activities such as Thomason et al., 2007 and Thomason et al. 2008). Comparisons of SAGE II with measurements by SAGE III and POAM III from the 2007 paper are shown in the figures below.

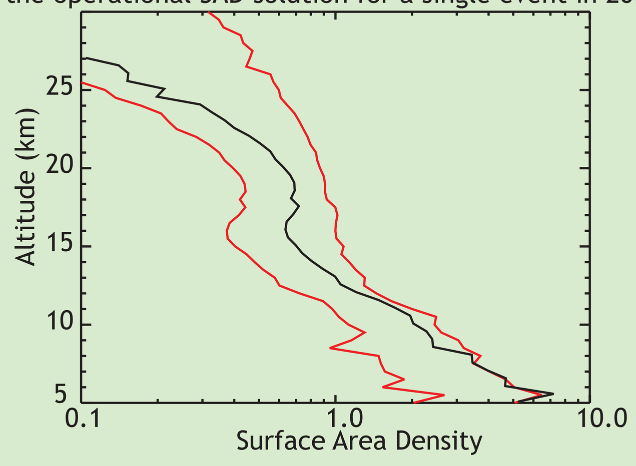
It is well known that visible and near-infrared extinction measurements are dominated by the largest aerosol present and small aerosol (<100 nm) are virtually invisible to these measurements. As a result, the derivation of bulk aerosol properties like SAD become increasingly bound to the retrieval methodology at lower aerosol loading where small aerosol become increasingly important. While not mathematically required to do so, most commonly-used methods are susceptible to preferentially underestimating SAD. Even using the well-accepted log-normal size distribution tends to yield very narrow, low number density size distributions with concomitantly low SAD values.

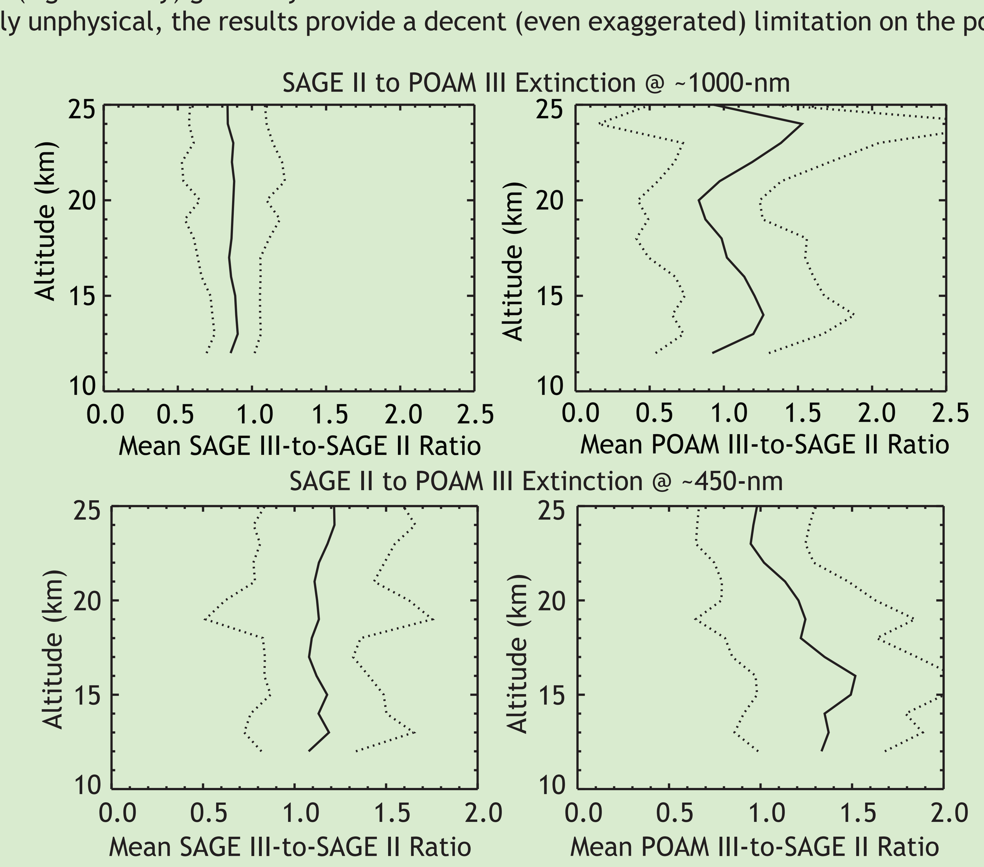
Recently, Thomason et al. (2008) examined SAD extremes that are consistent with the SAGE II extinction coefficient measurements. Unlike most methods, their approach limited the aerosol size distribution only by the total number of particles present and purposefully sought the extreme values of SAD. Ultimately, this method produced a pair of delta functions which controlled the observed wavelength dependent extinction and allowed either a minimal number of particles required to match the extinctions (as low as 1 cm^{-3}) or forced a relatively high particle number density (20 cm^{-3}). Under background conditions, they observed a range of a factor of 2 to 4 from the minimum to the maximum possible SAD as shown in the example profile figure below. This difference is (significantly) generally exceeds the observed differences between OPC and SAGE II-derived SAD. While the implied size distributions are grossly unphysical, the results provide a decent (even exaggerated) limitation on the potential for bias in SAGE II SAD estimates.

Thomason, L. W., Poole, L. R., and Randall, C. E., SAGE III aerosol extinction validation in the Arctic winter: comparisons with SAGE II and POAM III Atmos. Chem. Phys., 7, 1423-1433, 2007, www.atmos-chem-phys.net/7/1423/2007/.

Thomason, L. W., S. P. Burton, B.-P. Luo, and T. Peter, SAGE II measurements of stratospheric aerosol properties at non-volcanic levels, Atmos. Chem. Phys., 8, 983-995, 2008, www.atmos-chem-phys.net/8/983/2008/.

Comparison of the extreme SAD solutions (red) with the operational SAD solution for a single event in 2000.





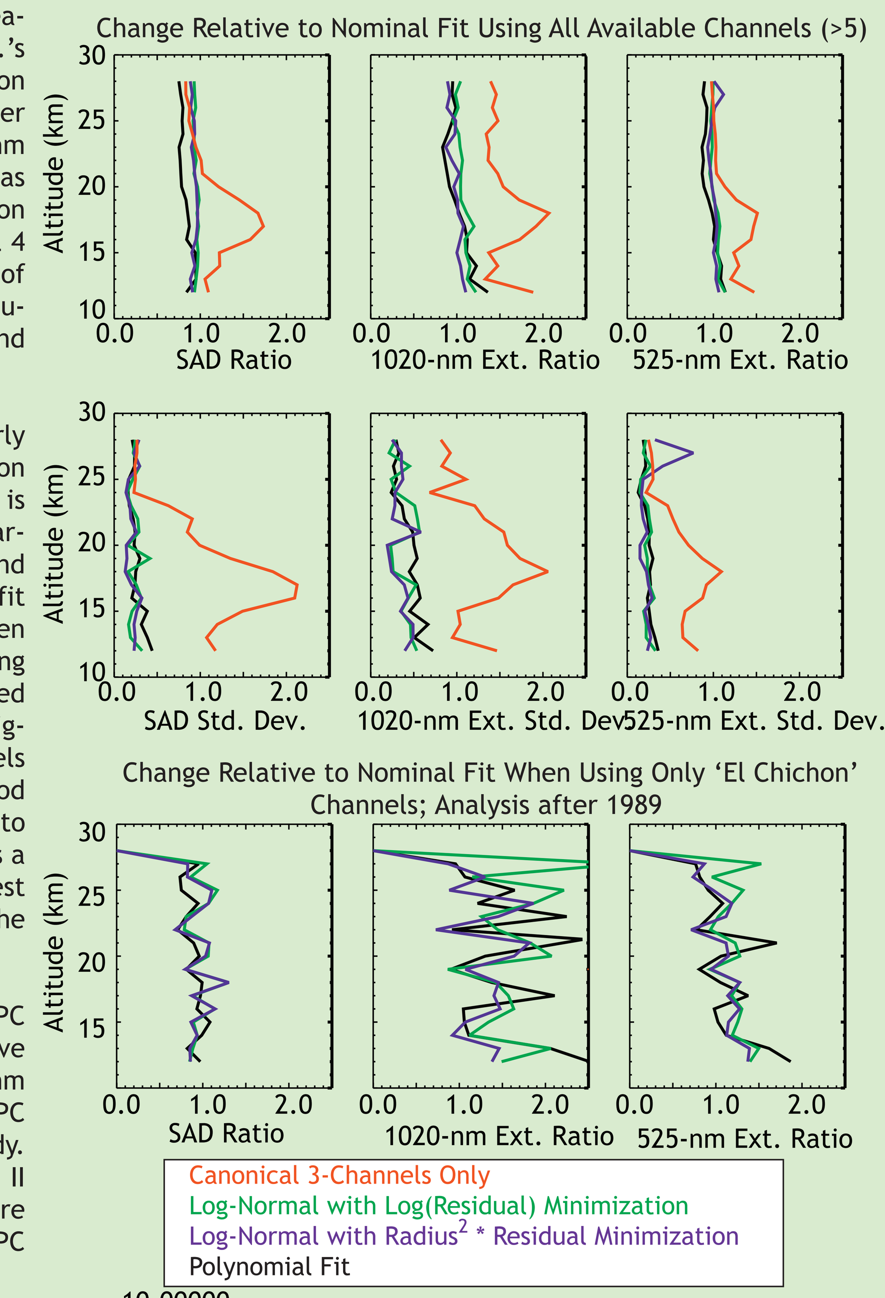
The University of Wyoming Optical Particle Counter

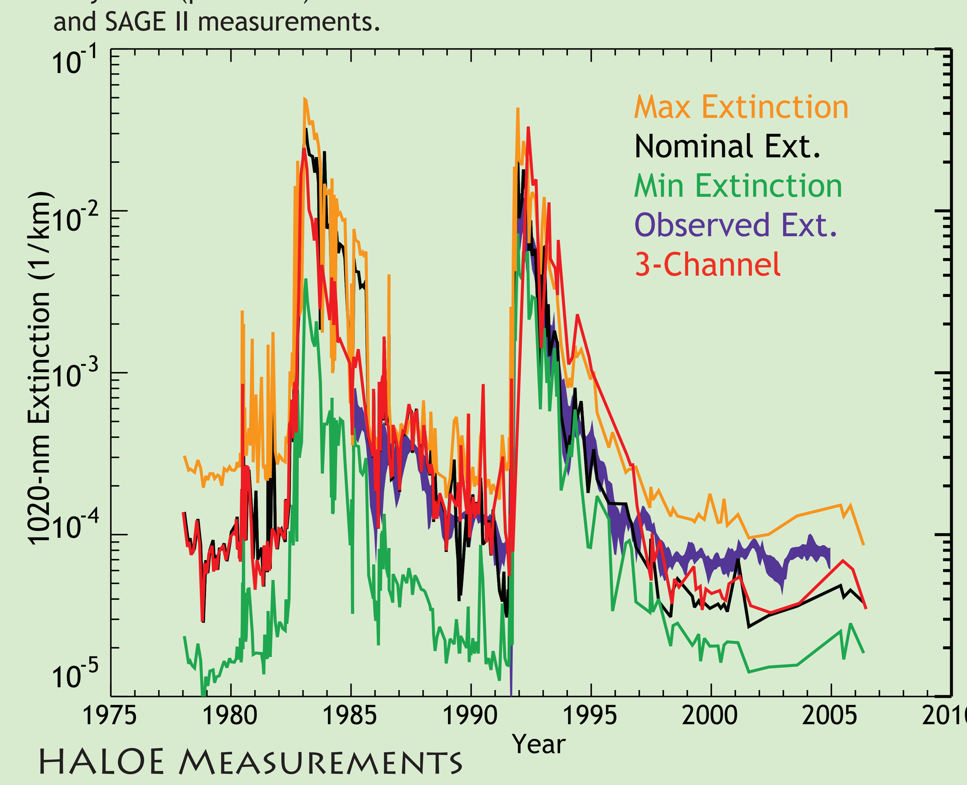
The University of Wyoming has collected data from Laramie (USA) since the mid-1970s. While the basic instrument function has remained unchanged, the number of size bins or channels has increased from 3 to as many as 13 over this time period. Below we examine the effect of the changing channel ensemble on the calculation of bulk properties such as SAD and 1020-nm extinction.

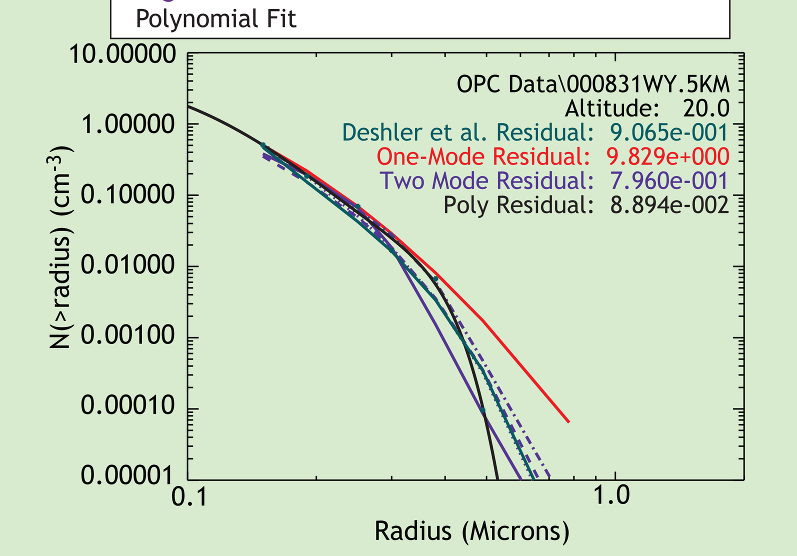
Initially, we produced a retrieval methodology that produced a single-mode log-normal fit. It can be used uniformly throughout the data record and always produces a viable fit whenever at least 3 channel measurements are available. This is applied to the 'canonical' channels: CN (0.01 μm), 0.15 and 0.25 μm. Since those channels are almost always available, we can produce a homogeneous (if less robust) time series without the impact of changing channel numbers. An example of the time series of 1020 nm calculated from the three channel solution and several other techniques is shown below. One interesting feature is that the 3-channel solution is typically less than Deshler et al.'s the bimodal/monomodal log-normal just after the El Chichon eruption whereas it is typically greater than the extinction computed just after the Pinatubo eruption. We have also developed a multi-mode algorithm that can use multiple initializations and minimization schemes as well as a solution based on a polynomial. The figures (right) show the comparison of Deshler et al.'s (2003) nominal size distribution with those from all 4 solution approaches developed here (restricted to when the number of non-zero channels is greater than 5). Note that both the multi-mode solutions developed here and that of Deshler et al. can produce null second modes.

The multi-mode solutions and the nominal solution tend to agree fairly well. The SADs are about 10% lower with an RMS of about 25%. Extinction values are typical closer within +/-20% but the standard deviation is greater at 25 to 50%. On the other hand, the single mode solution particularly below 22 km can be different by as much as 75% for SAD and 100% for 1020-nm extinction. This seems to follow the inability of the fit to produce a typically steep positive shoulder with a single mode driven by the smaller size bins (see the figure in the lower right) thus producing aerosol at larger sizes than is actually observed by the more advanced OPC. Another interesting feature is shown in the lower set of three figures on the right. Here, we use Pinatubo observations but only channels that were available (as close as possible) during the El Chichon period (CN, 0.15, 0.25, 0.95, 1.20, 1.80 μm). The SAD is not overly sensitive to the change in channels. However, aerosol extinction at 1020-nm shows a consistent increase to a mean of approximately 50%. This may suggest the reason for the reversal of the 3-channel solutions relative to the nominal solution.

At this point, we see good stability in the SAD measurements for the OPC when more than 5 channels are available. On the other hand, we have seen nothing to suggest a solution to the differences with SAGE II 1020 nm extinction. As a test, we have developed an extreme solution for an OPC size distribution that is analogous to that used in the SAGE II SAD study. The extreme solution, while non-physical, does encompass the SAGE II measured 1020-nm extinction measurements. This suggests that there may be a (palatable) size distribution that is consistent with both OPC and SAGE II measurements.







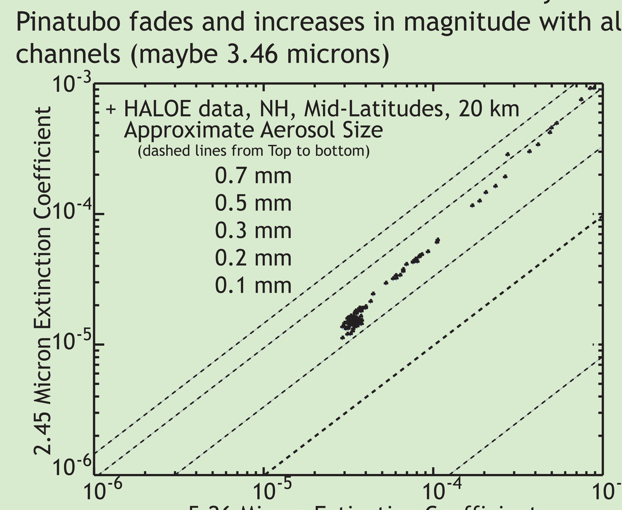
Deshler, T., M. E. Hervig, D. J. Hofmann, J. M. Rosen, and J. B. Liley, Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41°N), using balloon borne instruments, J. Geophys. Res., 108(D5), 4167, doi:10.1029/2002JD002514, 2003.

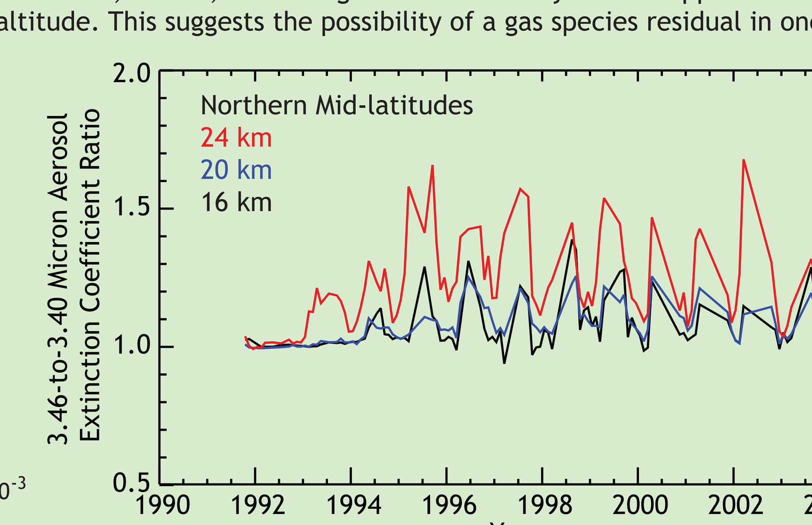
Wagner, R., A. Mangold, O. Mohler1 H. Saathoff, M. Schnaiter, and U. Schurath, A quantitative test of infrared optical constants for supercooled sulphuric and nitric acid droplet aerosols Atmos. Chem. Phys., 3, 1147-1164, 2003 www.atmos-chem-phys.org/acp/3/1147/.

HALOE Measurements

The HALOE analysis is in an early stage. We will continue this study and extend it to derived quantities such as SAD. So far, we have examined the internal consistency of the aerosol extinction data and found a few issues.

1. The 2.45 micron extinction coefficient data appears to be larger than possible for a nominally scattering-only wavelength (right). It could suggest that the refractive index information is in error but it does not seem possible for it be in error to the extent required by the observed anomaly (e.g., Wagner et al., 2003).
2. The 3.40 and 3.46 channels should be virtually identical but, in fact, show a significant annual cycle that appears as the effects of Pinatubo fades and increases in magnitude with altitude. This suggests the possibility of a gas species residual in one of the channels (maybe 3.46 microns)





Do models help clear the picture up?

Five aerosol modelling groups participated in the ASAP report. The models include both 2 and 3-D models and models that either produce their own aerosol in the entirety or require aerosol nuclei to be transported across the tropical tropopause. These include AER (Weisenstein), MPI (Timmreck), UMPC (Bekki), ULAQ (Pitari) and LASP (Mills). The figures below show the 1020-nm extinction and SAD profile results (relative to SAGE II) for these models under background conditions in northern mid-latitudes and in the tropics for April. They are designated by thin red lines and their mean is shown as a thick green line. Superimposed on these profiles are results for HALOE and the OPC (mid-latitudes) and HALOE alone for the tropics using only data after 1996.

For 1020-nm extinction, the models tend to produce more extinction (up to 50%) than is measured by SAGE II particularly in the lower stratosphere. This is in the opposite sense of the OPC/SAGE results (-50%) nor do they agree well with HALOE computed 1020-nm extinction. On the other hand, in mid-latitudes, the model mean and OPC SAD agree quite well while SAGE II results are a factor of 1.5 to 3 times smaller. The differences with SAGE II are due in part to the high fraction of the surface area density produced by models that reside in particles too small to be measured by SAGE II. The over-estimated extinction may suggest that the models similarly overestimate SAD, however, these quantities are not-linearly related and a more circumspect (and likely) conclusion is that closure has not yet been reached between measurements and the models. At this time, it is difficult to use model results to adjudicate the differences between SAGE II and the OPC. Nonetheless, it seems likely that space-based data sets particularly in the lower stratosphere under-estimate, perhaps significantly, aerosol surface area density.