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Atmospheric Measurement Techniques Discussions

Interactive comment on "A comparison of lognormal and gamma size distributions for characterizing the stratospheric aerosol phase function from OPC measurements" *by* Ernest Nyaku et al.

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General comments:
This manuscript deals with the nature of the particle size distribution of stratospheric sulfate aerosols. The main motivation is to improve assumptions on the aerosol scattering phase function required to retrieve aerosol extinction coefficients from satellite limb-scatter measurements. The study presents a re-analysis of balloon-borne measurements of the aerosol size distribution with optical particle counters. Specifically, two different size distributions (uni-modal log-normal and gamma distributions) are used to

model the observed cumulative distributions. The manuscript is interesting,presents relevant new information and should eventually be published in my opinion. The paper is very well written and generally easy to follow. There are several points I ask the authors to consider. Specific comments (often minor) are listed below. In addition, I have one more general comment:

The analysis is based on a more limited number of OPC channels than previous analyses of the measurements. In particular, the channels corresponding to large particle sizes are now not considered. These channels provided evidence for a second mode of the particle size distribution, even under background conditions. The second mode is now entirely neglected and the reader wonders, whether the authors now believe that the second mode does not really exist? I think this aspect should be explicitly addressed in the paper. The small number of large particles contributes substantially to the overall aerosol scattering signal and will probably also have a non-negligible effect on the scattering phase function. This is particularly relevant, because the gamma distribution systematically underestimates the number of particles for the largest size bin (top right panel of Fig. 6.)

Response: The fitting of two uni-modal distributions to OPC and OPC$_{nsb}$ data was motivated by Figure A2 of (Chen et. al 2018), who fitted four Bi-modal lognormal distributions (BMLN) to OPC$_{nsb}$ data measured on 12 April 2000 for altitude 20 km. All four fits had a similar AE of approximately 2.4, but each had different coarse mode fraction (CMF). These four BMLN distribution fits to the OPC data differed significantly from each other in the radius range between 0.01 µm to 0.1 µm and these differences resulted from the gaps in the OPC size bins that limited the ability of the fits to be constrained. All four fits captured the larger particles very well but the resulting phase functions differed from each other because of the overestimation or under estimation of the particles between 0.01 and 0.1 µm. The two gamma distributions are in good agreement with each other especially within the particle radius range of 0.01 µm to 0.1 µm but both systematically failed to capture the number concentration of the largest bin. Since the actual phase function is not known, it is possible that either of the phase functions from the UMLN or gamma may be right.

Also it should be noted that most of the OPC measurements associated with the OMPS measurement period lack measurable signals in the larger bins, especially in the 20-25 km altitude range that is most relevant for the current OMPS retrieval assessments (Chen et. al 2018). Therefore, these measurements do not provide a clear argument for the presence of larger particles in those cases. For cases that DO have a measurable signal for large aerosol bins, the signal remains much lower than in the smaller size bins. So (in a global, weighted fit like was performed), it may be appropriate for the fit to the data at the largest size bins to be very poor (relative to the the much higher signals in the smaller bins).

Specific comments:

1- Page2, line 31 "using Mie theory (Deirmendjian,1969)" I suggest citing the original paper by Mie here (Mie,1908)

Response: The line has been updated to "using Mie theory (Mie,1908)"

2- Page 2, same line "Here we make the assumption that the aerosol particles in the stratosphere are spherical" If Mie theory is used this assumption is implicitly made anyway. Perhaps this could be explicitly stated.

Response: This sentence has been revised to: "Theoretically, the $P_a(\Theta)$ is calculated from the aerosol size distribution (ASD) using Mie theory (Mie, 1908), generally assuming that the aerosol particles in the stratosphere are spherical and homogeneous."

3- Page3, line 61: "to correct the ASD"
Response: This sentence has been revised to read: "to retrieve the ASD"

4- Page3, line 66: "and found out that even if the particles were assumed to be spherical" I don't understand this part of the sentence, because, (a) if Mie theory is used the

particles are implicitly assumed to be spherical anyway, (b) if the HG phase function is used no explicit assumptions on the particle shape have to be made, right?

Response: The statement has been revised, it now reads: "Some techniques that have been used to model the $P_a(\Theta)$ are by computing it using the Henyey-Greenstein phase function (H-G) (Henyey and Greenstein, 1941; Ernst, 2013; Grams, 1981) or the modified Henyey-Greenstein phase function (MH-G) (Irvine, 1965; Cornette and Shanks, 1992) with a precise asymmetry factor g, which is the average cosine of the scattering angle weighted by the phase function. The shortcomings of using these functions to approximate the real Mie phase function were demonstrated by Toublanc (1996) for two cases. When the radius of the particle was ten times smaller than the wavelength, the H-G phase function failed to produce the shape of the real Mie phase function in comparison to that of the MH-G. By contrast, for a particle of radius that was comparable to the wavelength, both functions failed to reproduce the lobe patterns of the real Mie phase function."

5- Page4, lines108/109: coagulation is certainly also an important process for the growth of stratospheric aerosols.

Response: Coagulation is also an important stratospheric aerosol formation process and it has been included in the sentence, which now reads:

'A multimodal distribution can be used to represent coexisting "nucleation" , "coagulation", and "accumulation" modes after a volcanic eruption. The nucleation mode is associated with new particle formation from sulfur vapor which quickly coagulate to form larger particles (Hamill et al., 1997), and the accumulation mode associated with particle growth by condensation of the vapor on the existing particles (Steele and Turco, 1997).'

6- Page6, line 162 and equation(3): if O_i is already the "frequency" in each size bin, i.e. normalized to the total number of observations, then the multiplication of the expected probability values ζ_i with n in equation (3) is not required, is it? O_i corresponds to a probability then, and so does ζ_i

Response: O_i is the "frequency" in each size bin. It is normalized by dividing by "n". Rearranging the equation will then lead to the "n" in the denominator being squared (which was omitted). Equation (3) has now been updated squaring "n" in the denominator. On the other hand, if O_i is defined as the normalized "frequency" in each size bin, then the multiplication of the expected probability values ζ_i with "n" in the equation would not be required.

7- Page7, line 185: "The LPC data consists of 20 months of measurements" Table 2 lists more than 20 months.

Response: This sentence has been corrected to read: "The LPC data consists of 27 months of measurements" to correspond to the number of months listed in Table 2.

8- Page,10 Figure 1: I think it would be quite interesting for the reader to see plots of the non-cumulative versions of the gamma and UMLN distributions for these cases.

Response: Figure 1 has been updated to include non-cumulative versions of the gamma and UMLN distributions for the two cases shown.

9- Same Figure: It is also worth mentioning in the text that the ASDs differ substantially for radii > 300 nm. At 600 nm or so the difference one order of magnitude.

Response: The text has been updated to include the above suggestion:

"The two ASDs tend to diverge beginning at radii greater than 300 nm and differ substantially at approximately 600 nm, where aerosol concentrations are below the minimum detectable concentrations, and there these differences can reach one order of magnitude."

10- Page 11, line 242: "This is shown in Figure 5, where one observes a considerable change in the magnitude of the phase function, especially in the back-scattering directions ($\Theta \geq 90°$) for this X value" I don't think this statement is correct. Looking

at the Figure, the phase function for X=1 is almost constant for scattering angles > 90 degrees. Perhaps you intended to make another point?

Response: The above statement has been rephrased to read:

"The phase function for X = 3 shows a forward peak and is nearly constant for scattering angles ($\Theta \geq$ 70 °). When there are no measurements between the 0.01 and 0.15 µm bin sizes, then the particle concentration within this range is estimated by the function used to fit the data. Errors in estimating the number of particles within this range by the function used for fitting the data will lead to uncertainties in the phase function as shown by the X = 1 plot in Figure 5.

11- Page 12, caption Figure 5: "increase .. complexity of the phase function" The complexity (e.g. for X=10) is mainly due to the fact that a mono disperse aerosol is assumed here. If you assumed a UMLN or a gamma distribution then the oscillations will be damped.

Response: The caption of Figure 5 has been revised to read: "Mie phase functions of a monodisperse aerosol for different values of the size parameter X derived with a refractive index of 1.33. The increasing asymmetry and complexity (e.g. for X=10) of the phase functions with increasing X is due to the use of a monodisperse aerosol. The oscillations observed are damped when the phase functions are computed for an ensemble of aerosols that are assumed to have a UMLN or gamma distribution. The phase functions are shown for the range of scattering angles that are observed by OMPS, SCIAMACHY and OSIRIS."

12- Page 13, Figure 6 and related discussion in the main test: I certainly agree that the differences between the OPC-like and LPC-like fits are smaller for the gamma distribution than the UMLN distribution. However, both gamma distributions systematically underestimate the number of in the largest bin. If larger bins would be considered this low bias would probably be even larger. So the two gamma distributions are in good agreement, but they are also both systematically wrong. Perhaps their phase functions deviate even more from the actual phase functions compared to the phase function based on the UMLN distribution? Looking at the χ^2, the UMLN distribution without the extra measurement still show the best performance. I am not asking for any more analysis here, but I think it should be clearly stated that the gamma distributions fail to capture the OPC measurements for the largest sizes, which will lead a systematic error in the derived phase functions.

Response: "OPC-like" has been changed to OPC$_{nsb}$ and "LPC-like" has been changed to OPC: "nsb" stands for no small bin .

There was no arbitrary decision to ignore measurements from the largest bin when the fits were made. Thus the systematic underestimation of the concentration of the largest bin by the gamma distribution fits was not deliberate as the same can be seen for the UMLN fit (red line). Because the actual phase function is not known, it is possible that the phase functions derived from the gamma or the UMLN distribution functions may be the right one.

This has been stated in the text as “The failure of both gamma distributions to capture the OPC measurements for the largest bin size for the case shown in Figure 6 could lead to a systematic error in the derived phase functions.”

13- Page 14, line 287: "The gamma distribution does not have the same tendency to over estimate the larger particles" This is now different from the earlier analysis of the OPC/LPC measurements, where the gamma distribution systematically underestimated the large particles.

Response: These are the CARMA microphysical model outputs at Wyoming and these are different from the OPC/LPC measurements made at the same location.

14- Page 16, Figure 7: "The blue data points" I can’t identify blue points on my printout.
Response: The size of the blue dots has been increased.

15- Page 18, line 325: "Additionally, it has been shown that whenever OPC-like con-

centration measurements are made, the gamma distribution is the best distribution to be fitted" I don’t agree with this statement, because χ^2 for the OPC-like measurements is significantly smaller for the UMLN distribution than the gamma distribution. Please rephrase this statement to eliminate this apparent contradiction. As mentioned above, the difference between the gamma-fits for the OPC-like and LPC-like measurements are admittedly very small, but the gamma distribution systematically under estimates the measurements for radii > 300 nm. Since the large particles dominate the scattering signal, they will have a non-negligible effect on the phase function. It may even be possible that the OPC-like UMLN distribution yield a phase function that agrees best with the actual phase function.

Response: Figure 6 is a comparison of the fits made with the inclusion of a small bin (OPC) to one with no small bin (nsb) OPC$_{nsb}$ measurements. On this figure, the χ^2 value for OPC UMLN distribution fit is 0.0135 and that of gamma distribution fit is 0.0101. This shows that the χ^2 for the gamma fit is somewhat less than that of the UMLN fit. This is in agreement with the statement:

"Additionally, it has been shown that when the same LPC concentration measurements are fit without using the 0.092 μm bin, the gamma distribution provides a some what better fit because of its insensitivity to particles between 0.01 μm and 0.1 μm range when compared to the UMLN distribution; however the gamma distribution in both cases underestimates the concentrations of the larger particles, which may be quite important depending on the wavelength of interest"

16- Page 18, general comment on the conclusion: the 2nd mode reported in earlier study is now entirely neglected. The earlier OPC measurements showed indications for the second mode even under background conditions. I guess this measurements are still valid- they are also based on more channels at larger radii. It would be good if the authors would comment on how to treat the second mode in future studies. The large particles with radii of several 100 nm may have a substantial impact on overall scattering properties and the phase function of stratospheric sulfate aerosols.

Response: During background conditions, Deshler et al. (2003) have in some cases used a bimodal lognormal (BMLN) particle size distribution to achieve the best fit to the OPC measurements made by the in situ optical counters. However, with limb scattering geometry this BMLN size distribution is not possible because six (or five when the data points are normalized) independent pieces of information at each altitude will be needed to describe the BMLN distribution, but at altitudes greater than 20 km, OPC measurements mostly provide four data points. This makes it impossible to fit a bimodal distribution.

Also, we agree that particles with radii larger than 100 nm may have a substantial impact on the overall scattering properties and the phase functions of the stratospheric sulfate aerosol during volcanically active or periods with pyro CB activity. But most of the limb radiance measurements made by OMPS in the last seven years is devoid of any large volcanic activity sufficient enough to inject aerosols into the stratosphere and are mostly considered as background condition. In the future, we hope to compare the phase functions derived from multi-modal aerosol size distributions.

Typos etc,:
1- Page 2, line 7: "Philippines,1991" -> "Philippines, 1991"
Response: This has been corrected.

2- Page 2, line 35: ",longitude" -> ", longitude"
Response: This has been corrected.

3- Page 3, line 58: "occulation" -> "occultation"
Response: This has been corrected.

4- Page 3, line 58: "was began" -> "was begun"
Response: This has been corrected.

5- Page 3, line 59: I think "that" in "that provided" can be omitted.
Response: This has been corrected.

6- Page 3, line 67: "calculations .. was" -> "calculations .. were"
Response: This has been corrected.

7- General comment on spelling of "Angström": sometimes you use "A" as the first letter,sometimes "Å". I think the latter is correct and should perhaps be used throughout the manuscript.
Response: This has been updated everywhere in the manuscript.

8- Page 3, line 75, equation (1): "nm" can be omitted here (4 occurrences)
Response: This has been corrected for all occurrences.

9- Page 4, line 86: "on measurements from Laramie, Wyoming optical particle counter (OPC) measurements"
Response: This sentence has been corrected to read "on data from Laramie, Wyoming optical particle counter (OPC) measurements"

10- Page 4, line 98: "by (Deepak .." and next line "or (Hinds" Wrong cite command used (\citep -> \cite}
Response: This has been corrected.

11- Page 4, line 10: "Sparc" -> "SPARC"
Response: This has been corrected.

12- Page 7, line 183: "following (Kovilakam" \citep -> \cite
Response: This has been corrected.

13- Page 9, line 226: add space after "shape parameter"
Response: This has been corrected.

14- Page 12, line 260: ".This" -> ". This"
Response: The correction has been made.

15- Page 14, lines 264 and 266: \citep -> \cite
Response: This has been corrected.

16- Page 18, line 309: "in the along the" -> "along the"
Response: The correction has been made.

17- Page 23, line 471: "Sparc" -> "SPARC"
Response: The correction has been made

18- Same line: add space in "(eds.),SPARC"
Response: The correction has been made.