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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Interactions of meteoric smoke particles with sulphuric acid in the Earth's stratosphere" *by* R. W. Saunders et al.

**Anonymous Referee #2**

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This paper addresses a gap in our understanding of the interaction of meteoritic smoke particles (MSPs) with gas-phase sulfuric acid at altitude above the top of the stratospheric sulfate aerosol layer. It attempts to explain observations of decreasing sulfur-bearing H2SO4 (and SO3, though not mentioned here) gas that have long been surmised to be due to interactions with MSPs, but which are in need of this type of detailed examination.

The paper is divided into three sections: laboratory studies of the acid dissolution of materials with the surmised MSP composition, atmospheric modeling studies of MSP growth and transport, and comparison of model calculations to observations. I am not qualified to comment on the methods used in the laboratory studies. The modeling studies are significantly more primitive than previous studies by Megner et



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al. (2008) and Bardeen et al. (2008), and differences between these and the previous calculations suggest significant limitations of the usefulness of this new model for this study, undermining this study's conclusions. The comparisons to observations further suggest weaknesses in the model.

Comparison of Figure 4 to the vertical profiles of number density calculated by both Megner et al. and Bardeen et al. reveal that something is very seriously different about this new model calculation. Both of these previous studies used sophisticated bin microphysical codes that included sedimentation and coagulation, and used a constant flux of meteoritic material as a source function. In contrast, this study treats MSPs as a gas-phase tracer which neither sediments nor coagulates, and sets a constant concentration of MSPs at the 80-km upper boundary of UMSLIMCAT.

The authors deal with the lack of sedimentation and coagulation by claiming that Bardeen et al. showed that sedimentation is relatively unimportant in their calculations. This may be true for particle number, but it is certainly not true for mass. Figure 10 in Bardeen et al. shows increases in mass density exceeding 110% when sedimentation is turned off. Since MSP mass is likely the most important parameter in answering the questions about interactions with sulfates and H2SO4 gas, this model deficiency cannot be brushed off.

Moreover, the vertical profiles of number densities for UMSLIMCAT shown in Figure 4 look nothing at all like those shown in Bardeen et al., or in Megner et al. These previous studies produce number density profiles similar in shape and magnitude to the red curve in Figure 4, for the 1D calculations which include coagulation. They show decreases at altitudes below the 80-km peak number density, with significant latitudinal and seasonal variability. In contrast, UMSLIMCAT produces number densities that increase monotonically with decreasing altitude.

The authors chose to specify a constant number concentration of MSPs at 80 km, rather than a constant flux, as Bardeen et al. and Megner et al. did. It is reasonable to



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expect that there is a somewhat constant flux of meteoritic material entering the Earth's atmosphere at all times and latitudes (though studies by Janches and others indicate there is some seasonal and latitudinal variation even to the flux). But as both previous studies show, a constant flux does not produce a constant number concentration of MSPs at 80 km, the peak source altitude. This is because strong winds advect the particles latitudinally, and cause them to diverge from the summer pole, and converge at the winter pole.

Figure 5 shows contours of mass density for MSPs that are oddly seasonally asymmetric. In contrast, the contours shown in Megner et al. and in Bardeen et al. show a seasonal symmetry to the hemispheres that seems much more reasonable. The authors do not mention this troubling result from their model.

The comparison to SOFIE observations in Figure 6 show 2 vertical profiles with very different slopes, in contrast to the comparisons in Hervig et al. (2009, Fig. 2) with calculations provided by CHEM2D (Megner) and WACCM (Bardeen). This is another indication that the vertical distribution of MSPs calculated here has serious deficiencies compared to previous work. As Hervig et al. shows, the magnitude of MSP extinction can easily shift by 2 orders of magnitude depending on the assumed composition of the MSPs. But the slope of the profile remains the same. Hence the fact that the 2 profiles in Figure 6 intersect at 45 km means little, as the values could easily shift for a different assumed MSP composition. But the difference in slope between the 2 profiles is significant, indicating a strong lack of agreement with SOFIE and with previous models.

The treatment of H2SO4 in this study appears to be extremely simplified, with no chemical reactions, phase changes, or photolysis indicated in the manuscript. Still, this simplification may be reasonable for the purpose here of dealing with the fate of H2SO4 above the top of the stratospheric sulfate aerosol layer, as it interacts with MSPs at higher altitudes. Figure 7 shows possible agreement with the 2 observations of Arijs et al. (1985) at 45 km. Yet Figure 7 fails to show a third observation from the very same



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paper at 42.5 km. Why has this been left out? Given the paucity of observations at this altitude range, it is important to include all available data. It appears that the model does not agree as well with this missing data point.

I suggest one of two possible remedies for the deficiencies in the model used in this paper. The authors could significantly improve their model calculations of MSP distribution to better match observations and previous, more sophisticated calculations. This would involve including sedimentation, coagulation, and a constant flux of meteoritic material. Alternately, they could remove their MSP calculations from the manuscript and use the calculations presented in Megner et al. and Bardeen et al. as inputs to their own calculations of the interactions of MSPs with H2SO4. Based on their laboratory measurements and these sectional models, one could derive the required uptake coefficients to account for observations with much greater confidence.

Technical Corrections

Page 1555, Line 18: I believe the reference should be “Neely et al., 2011”, as the suffix “III” to Neely’s name should not be included.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 1553, 2012.