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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.

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This referee has commented only on the modelling aspects of the paper, stating that they did not feel qualified to comment on the experimental part of the study. Our responses to the points raised in this review are as follows;

• Model validity

We first owe an apology to the referee, who was understandably concerned about the asymmetry in Figure 5 (which shows the distribution of meteoric smoke particles (MSPs) as a function of latitude at four different seasons). The apparent asymmetry resulted from contouring the data with too coarse a latitudinal resolution (10 degrees).

The figure panels have thus been re-plotted at higher resolution (1 degree). We trust, given the similar general features of the MSP distribution in the stratosphere to the previous studies of Bardeen et al. (Figure 3) and Megner et al. (Figure 3), that the referee's concern is now allayed. The model data which was used for determining the meteoric loading of stratospheric sulphate aerosol (SSA), and removal of sulphuric acid in the upper stratosphere, is of course unchanged.

The referee states that our modelling approach is "significantly more primitive" compared with the previous studies of Bardeen et al. and Megner et al. While it is correct that we do not include sedimentation and coagulation of MSPs, the important point is that the additional complexity in those earlier studies does not make any significant difference to the modelled *volumetric smoke mass* in the stratosphere (this is discussed in more detail below). Furthermore, our volumetric surface area is valid for obtaining the lower limit to the acid uptake coefficient on smoke particles. **Volumetric mass and surface area are the two quantities we required to meet our primary objectives, which were to assess the impact of MSPs on SSA in the lower stratosphere and acid vapour in the upper stratosphere respectively**

• Sedimentation/coagulation/constant mass flux

The referee states that the previous multi-dimensional studies differed from our approach by using sophisticated treatments of sedimentation and coagulation. Firstly, we note that these sedimentation and coagulation treatments are essentially no different to those used in the original 1-D modelling study of Hunten et al. (1980) i.e. over three decades ago, and so in themselves do not represent a significant advance merely through implementation in a 2/3-D model. Indeed, we (Gabrielli et al, *Nature*, 432, 2004; Saunders et al., 2007) and others have used similar coagulation/sedimentation schemes in the intervening years when considering possible MSP size distributions in the middle atmosphere.

Regarding sedimentation, Figure 10 of Bardeen et al. indicates to us that over most of the relevant altitude range of UMSLIMCAT (80 km and below), including sedimentation would have no significant impact on the modelled MSP mass. As those authors point out, in the stratosphere the difference is less than 30%, which is below the level of significance. As stated in our paper, initial runs of UMSLIMCAT with coagulation incorporated using an equivalent particle size bin method (Saunders et al., 2007) showed that there were no significant differences to the runs without coagulation. We find the same result with our 1-D model (Figure 4 in the paper).

Regarding coagulation, the first point to bear in mind is that, to the best of our knowledge, there is actually no direct evidence that coagulation of MSPs occurs in the atmosphere. There is good evidence from rocket-borne charged particle detectors and incoherent scatter radar analysis that small primary MSPs (radius 1 - 2 nm) form above 80 km (Rapp et al., 2007), but conjectures about when and how coagulation then occurs are based on laboratory studies in our group (Saunders and Plane, 2006; 2011). Almost all previous MSP models have assumed that coagulation occurs solely through Brownian motion-collisions which maintain particles of spherical shape and compact structure. In fact, in our laboratory studies where analogue MSPs consisting of Fe-Mg silicates were synthesised, fractal-like morphologies were consistently observed and explained by the role of magnetic dipole-induced aggregation of Fe-containing particles. We have shown previously (Saunders et al. 2007) that magnetic dipole-induced aggregation can reduce the particle number in the upper stratosphere by over an order of magnitude. However, there is a further point to consider: in the atmosphere these particles form and grow in a huge excess of water vapour (approx. 1e+05 times greater concentration), so that hydroxylation of the particles may reduce the magnetic-dipole driven coagulation rate, as well as the degree of porosity of the particles. Weathering by H_2SO_4 in the upper stratosphere may have similar effects. We conclude that there has to be significant room for speculation regarding the accuracy of the MSP numbers reported in the Bardeen et al. and Megner et al. studies – this is an important point acknowledged in both these papers.

The referee refers to the profiles in Figure 4 of our paper, stating that the 1-D profile which includes coagulation is consistent with the Bardeen/Megner profiles but the UMSLIMCAT profile (where coagulation is not treated) exhibits, in contrast, a monotonic increase at lower altitudes. Of course, such a difference in the height-resolved number distribution is expected when a treatment of coagulation is included. The good agreement between our 1-D model-derived profile (accounting for coagulation/sedimentation) above 80 km and that from the cited 2/3-D models (and the model of Hunten et al) is what gives us confidence in using the numbers generated from the 1-D model as an input at the top level of UMSLIMCAT.

In terms of the meteoric mass flux, the previous modelling studies have used a base case of 44 tonnes of interplanetary dust per day. This number dates back to a meteor radar extrapolation by Hughes, and was a figure widely quoted in modelling the mesospheric metal layers up until a few years ago. However, recent radar measurements and modelling studies (discussed in Vondrak et al., 2008) indicate that this is very likely to be an overestimate. The seasonal variations in meteoric flux to the atmosphere referred to by the referee were studied by Megner et al., who concluded that this factor has a minor effect on predicted MSP numbers in the stratosphere. We also looked into this using a meteor input function from Janches (see Gardner et al., *JGR*, 116, 2011), and reached the same conclusion: while the seasonal meteor input function affects the metallic layers in the upper mesosphere, these effects "wash out" because of the many months taken by MSPs to reach the stratosphere.

The referee suggests using the MSP volumetric masses published in the Bardeen et al. and Megner et al. studies for our estimate of the H_2SO4 uptake coefficient. There are two points to make here. First, those studies do not actually agree particularly well. For an identical meteoric flux of 44 tonnes per day, Figure 3 in Bardeen et al. shows higher volumetric masses by a factor generally around 2-3 compared with Figure 3 of Megner et al., although there are some areas in the polar and mid-latitude stratosphere where the disagreement is more pronounced. Second, in order to investigate H_2SO_4 uptake

on MSPs in the upper stratosphere, we needed to include a self-consistent treatment of the downward transport of MSPs from the mesosphere *together with* upward transport of H_2SO_4 from the lower stratosphere, within the same model.

We used a constant MSP number concentration at 80 km (derived from our 1-D model), consistent with rocket-borne charged particle measurements (Lynch et al/Gelinas et al) above 80 km. We then showed that this input produced a SSA Fe Wt% in good agreement with aircraft measurements at 20 km. We argue that this provides a fairly tight constraint for UMSLIMCAT.

• SOFIE comparison

We consider that the level of agreement between the altitude profile of the SOFIE extinction at the single reported wavelength in Figure 2 of Hervig et al (2009), and the fitted profiles using the WACCM and CHEM2D-derived particle numbers and an assumed single composition, is somewhat fortuitous. This is because of uncertainties in: (i) the particle shape/size/composition; (ii) the precise nature of coagulation mechanisms; (iii) the use of bulk crystalline refractive index data applied to amorphous nanoparticles, and (iv) the likely evolution of the optical properties of MSPs as a result of chemical processing by water and acid vapours. It therefore seems unlikely that the good agreement can realistically be put down solely to a rigorous model treatment of all relevant processes and parameters.

In order to make this point more explicit, we have now added to Figure 6 the extinction profiles shown in Hervig et al., which were calculated from the WACCM and CHEM2D studies assuming a *pyroxene* composition for particles. Hervig subsequently (COSPAR General Assembly, Bremen 2010) showed that additional wavelength analysis from SOFIE data indicates that an *olivine* composition for MSPs is more likely, consistent with our laboratory study (Saunders and Plane, 2011). Figure 6 now shows that *none* of the three studies actually matches the SOFIE observations over the entire height

range from 40 to 75 km. CHEM2D does best above 50 km, WACCM below 50 km, and UMSLIMCAT around 50 km. In our paper, we discuss the various factors (such as fractal geometry and porosity) which are most likely to account for the deviations between the SOFIE data and the calculated UMSLIMCAT extinction profile. The important point here is that the level of agreement between the SOFIE and UMSLIMCAT extinction profiles, given the uncertainties listed above, provides evidence that the modelled MSP mass in the stratosphere is correct within a factor of 4.

The difference in the slope of our data in Figure 6 compared with the other models results from differences in the number density profiles, which obviously depend on whether coagulation is treated or not.

• Sulphuric acid uptake

In our study, we were interested in ascertaining the likely impact of MSPs on stratospheric gas-phase H_2SO_4, in terms of the heterogeneous uptake of the gas onto available particle surface area. Subsequent chemical reactions or phase changes at particle surfaces referred to by the referee, and the separate loss of acid by photolysis, have been assessed in previous modelling work (e.g. Mills et al., 2005) and will all occur on a much slower timescale than heterogeneous uptake, and should therefore be less significant.

The data point and associated error bar at 42.5 km in Figure 7 have been added, as suggested. Their inclusion only further supports our conclusion that the uptake coefficient would need to be at least 0.01 to account for the observed decrease in acid above 40 km. Note that if coagulation is taken into consideration regarding acid uptake onto MSPs, the consequent reduction in available surface area would require a higher uptake coefficient (i.e. consistent with our lower limit of 0.01).

• Technical correction

Citation corrected as suggested.

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