

Interactive comment on “Interactions of meteoric smoke particles with sulphuric acid in the Earth’s stratosphere” by R. W. Saunders et al.

Anonymous Referee #1

Received and published: 28 January 2012

Interactions of meteoric smoke particles with sulphuric acid in the Earth’s stratosphere
Saunders et al. ACPD-12-1553-2012

This manuscript has three somewhat independent sections. It describes laboratory experiments on the dissolution of Mg-Fe-Si minerals relevant to ablated meteoric material in sulfuric acid. It also presents model results of the transport of meteoric smoke particles in the stratosphere. Finally, it compares some estimates of the extinction of meteoric smoke with data from SOFIE.

The laboratory experiments are useful, showing that test tube experiments with crystalline materials overestimate the time for smoke particles to dissolve/react in sulfuric acid. They also show oxidation of the iron. However, the experiments seem incomplete. First, there are no standards shown for calibration of the UV spectra. The Fe

C34

peaks shift significantly in both wavelength and strength with acidity, sulfate ion concentration, and other factors. I’m not clear that the literature spectra cover the range of acidity and temperature studied in this manuscript. Second, the solubility was not actually measured even though it should have been straightforward to at least get an estimate. On page 1560 there is just a rough calculation based on literature absorption values. Why not add different amounts of olivine powder to sulfuric acid and see where the Fe(III) absorption saturates (after sufficient time for dissolution)?

A significant question relevant to the stratosphere is that there is no information on the fate of the Mg and Si. The conclusions state “meteoric smoke particles will fully dissolve in the stratospheric aerosol layer within a week” (p. 1571). In fact the data do not prove this – the UV spectra only prove that the Fe dissolves, not that the Si and Mg dissolve.

The model results are very weak. The omission of sedimentation in this manuscript is much more important than the authors suggest. Sedimentation is only unimportant in the upper stratosphere. In the lower stratosphere, sulfuric acid condensation makes the particles large enough for sedimentation to be significant. Sedimentation is also significant in the mesosphere where the air density is very low. Finally, the statement on p. 1563 that sensitivity tests of Bardeen et al. concluded that sedimentation is of secondary importance is somewhat misleading. Their figure 10 shows roughly a factor of two changes due to sedimentation in many regions. Overall, the model results do not add to those previously published by Bardeen et al. (2008). The model also detracts from the next topic, the mass flux estimates.

The third topic is modeling of the extinction to obtain a flux of meteoric ablation and comparison to the observed iron to sulfur ratio by Cziczo et al. in the lower stratosphere. The results are obtained through Mie scattering and the UMSLIMCAT model. This topic is unnecessarily complicated. Mie scattering is not required at the accuracy of this manuscript. For particles much smaller than the wavelength, the extinction is proportional to the amount of absorbing material and its absorption cross-section.

C35

For nanometer particles, the absorption is essentially independent of the particle size distribution and shape. Using the model leads to misleading statements such as the end of p. 1566, where increasing the radius changes the extinction not because of a change in optical properties but simply because the mass goes up if the particles are bigger at constant number density.

Likewise, the iron to sulfur comparison is too complicated. Their ratio in the midlatitude lower stratosphere is essentially independent of the details of model tracer transport (e.g. Plumb and Ko, 1992). The ratio may depend on sedimentation, which is not included in this manuscript, and definitely on the assumed sulfur flux, which is not stated. So the statement in the conclusions that "modeling the MSPs from the upper mesosphere to the troposphere" is misleading: the match to iron content depends mostly on the assumed fluxes, not on modeling. The model only complicates the comparison.

Minor comments: - Figure 2 should mark the points where data were obtained. - I do not agree that "Figure 6 shows reasonable agreement" (p. 1566 line 23). There is over a factor of 10 relative discrepancy between 45 and 75 km. Given that the extinction depends mostly on the mass concentration, this means either the vertical transport is way off or there are large changes in refractive index with altitude. - page 1570 the comparison to HNO₃ is not very relevant because it is comparing reactive uptake of HNO₃ with sulfuric acid, which can undergo non-reactive uptake. A lower limit of 0.01 to the accommodation coefficient of sulfuric acid is not especially interesting

Recommendations: - The calculations of the meteoric flux should be simplified to show what assumptions are or are not actually needed to make the comparison with SOFIE and with Cziczo et al. The sulfur flux needs to be specified. - The model results should include sedimentation or be removed from the paper. Comparisons to the extinction profiles can probably be made by scaling the results from Bardeen et al. and Megner et al. - The authors should consider additional work on spectral standards and measuring solubility at relevant sulfuric acid concentrations.

C36

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

C37