

Interactive comment on “Chemical analysis of refractory stratospheric aerosol particles collected within the arctic vortex and inside polar stratospheric clouds” by Martin Ebert et al.

Anonymous Referee #1

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This manuscript describes electron microscope analysis of samples taken on high-altitude Geophysica flights in the Arctic stratosphere. The measurements seem carefully done. However, I think that the implied concentrations of large particles are on the edge of plausibility.

The manuscript describes 759 particles with mean diameter of about 1 μm obtained during 11 samples of 20 minutes each. The volume flow rate was 7.7 $\text{cm}^3 \text{s}^{-1}$; I verified that this is consistent with the stated orifice diameter. If I'm doing my math right the implied concentration is about 0.1 ppbm at the density of sulfate or about 0.2 ppbm at the density of silicates or aluminum oxide.

This concentration can be compared to measurements made in non-volcanic condi-

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tions by Deshler et al. and Wilson et al. A typical Wyoming balloon size distribution at 20 km had a mass mixing ratio of 0.35 ppbm, mostly in a small mode (Figure 5b, Deshler et al., 2003). The large mode in Deshler's figure was 0.02 ppbm, so the implied concentration for this manuscript would be at least 5 times as large. Even more relevant, Wilson et al. (2008) reported size distributions within the Arctic vortex at similar altitudes to this manuscript. The total aerosol concentration for relevant conditions was about 0.1 to 0.2 ppbv as sulfuric acid, or 0.4 to 0.8 ppbm (Figure 5 of Wilson et al., red points). For low N_2O (i.e. in the polar vortex), there were essentially no particles larger than about 0.8 μm diameter in the non-volcanic conditions (Figure 4 there).

It seems that the concentrations of particles larger than 0.5 μm implied in this manuscript are larger than previous measurements. The mass above 0.5 μm would be a significant fraction of the total aerosol mass in the Arctic stratosphere. The particles also appear to have minimal sulfuric acid coatings. This suggests a short residence time, which is consistent with sedimentation of large particles (>1 μm particles fall from the mesosphere to below 20 km in <6 months) but also implies a large source to maintain the concentration.

I can't think of a likely source. Globally, it is unlikely that there is enough mass in partially ablated meteoroids for such a concentration. I guess it is possible that the concentration in the polar vortex is much (factor of 10?) larger than in other parts of the stratosphere. Descent into the polar vortex might sweep particles from much of the mesosphere into the polar winter stratosphere, giving a higher concentration. I'm not sure this can be supported by other observations, though.

Even if there is enough partially ablated meteoroid debris, there is another issue. Because particles of about 1 μm diameter sediment out of the stratosphere several times as fast as smaller particles are removed and because the literature suggests that the majority of incoming meteoroid mass ablates, 0.2 ppbm of unablated meteoroid residue would imply a much larger concentration of meteoric smoke, probably over 1 ppbm. That would be inconsistent with Wilson et al.'s data and should have given large con-

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centrations of silicon, iron, and other elements on the second impactor stages in the samples here.

Other sources are less likely. The particles are spread over 11 flights so it is not a plume. The amount of ablating spacecraft debris is far too small. The composition does not match rocket exhaust: compared to the manuscript finding a lot of silicates, rockets produce far more alumina (solid fuel) or soot (kerosene fuel). The Sarychev eruption plume did not reach much above 15 or 18 km and was largely gone by the time of the RECONCILE samples (Jegou et al., 2013).

In summary, the manuscript describes what appears to be a reasonable analysis of data that lead to a large mass concentration, possibly plausible, possibly not. The authors need to consider the absolute concentrations and relate them to other literature. Wilson et al. (2008) is an obvious comparison. The authors could also calculate if the inferred concentrations of large particles should have shown up on lidar or other remote sensing observations. Whether this manuscript requires minor or major revisions depends somewhat on the results.

As long as there is no obvious conflict with other observations this analysis can be published with some caveats. I'm somewhat skeptical but don't want to stop results from being published when there is nothing obviously wrong with the technique.

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