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The Importance of Acid Processed Meteoric Smoke Relative to Meteoric Fragments for Crystal Nucleation in Polar Stratospheric Clouds

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Nitric Acid Trihydrate (NAT) crystal formation in the absence of water ice is important for a subset of Polar Stratospheric Clouds (PSCs) and thereby Ozone. However, nucleation of these crystals is not understood.

It has been suggested previously that either fragmented meteoroids or meteoric smoke particles (MSPs), or possibly both, are important as heterogeneous nuclei. The role of H_2SO_4, which is present in liquid PSCs, in these nucleation processes has not been investigated. It is known that metal-containing Meteoric Smoke Particles (MSPs) are processed, partially dissolving whilst some components re-precipitate within H_2SO_4 droplets, producing silica and alumina particles which differ in size from the original MSPs. We recently found that analogues for nanoparticulate MSPs have a low ability to nucleate NAT relative to larger particles of similar material, suggesting that the size of particles may be a critical parameter for the nucleating ability of silica particles. We previously showed experimentally that nano-particulate fumed silica is a poor promoter of nucleation, whilst micron scale fused quartz was found to be effective. Both materials have similar chemical and structural (crystallographically amorphous) properties.

In this study we developed a model using Classical Nucleation Theory (CNT) where we account for surface curvature of primary grains. This model is able to account for the discrepancy in nucleation effectiveness of fumed silica and fused quartz, by treating them as having the same nucleating ability (contact angle) but differing particle size (or equivalently surface curvature), assuming interfacial energies which are physically reasonable given literature measurements. We also performed new experiments which allowed us to refine our understanding of the H_2SO_4 sensitivity of NAT nucleation by meteoric fragments. Combining sedimentation modelling with our results and recent experiments on fragmentation of incoming meteoroids suggests that fragments are unlikely to be important as heterogeneous nuclei. However, the CNT model developed here provides evidence that nucleation of NAT on (10s nm) MSP analogues is effective enough to explain observed NAT crystal number concentrations in PSCs (without ice).