Supplementary material

XRD of SiO_2

X-Ray Diffraction (XRD) patterns of the two SiO_2 materials are shown in Figure S1. No crystalline phases were detected.

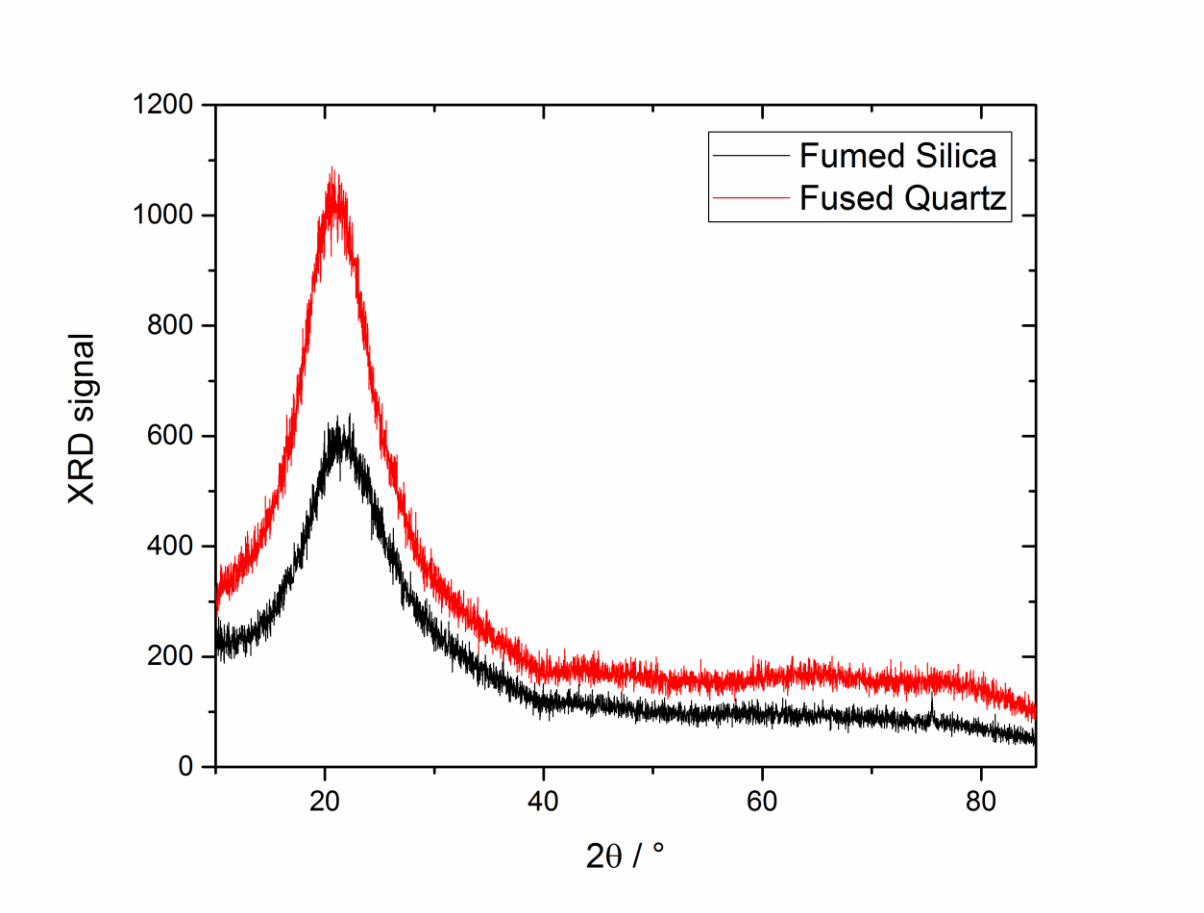

Figure S1: Powder X-Ray Diffraction (XRD) of SiO_2 materials. Diffraction patterns for the two SiO_2 materials, showing broad peaks from the amorphous materials, but no sharp peaks from crystalline phases.

Electron Microscopy of SiO_2

Fused quartz and fumed silica were both imaged in a Scanning Electron Microscope (FEG-SEM – FEI Nova 450) and the elemental composition of the fused quartz examined with Energy Dispersive X-ray spectroscopy (EDX, AMTEK at 18 kV). Figure S2 shows SEM images of the two SiO_2 materials.

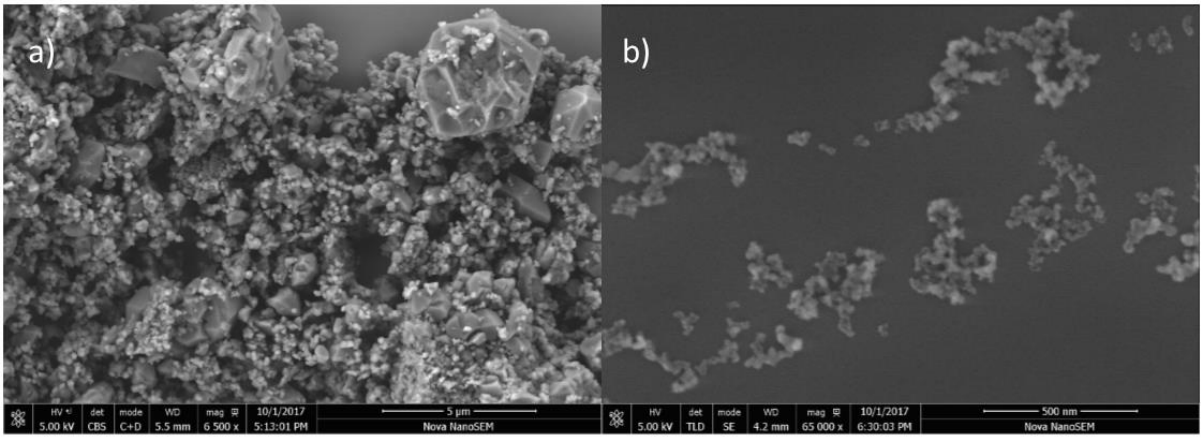

Figure S2: SEM images of a) fused quartz and b) fumed silica.

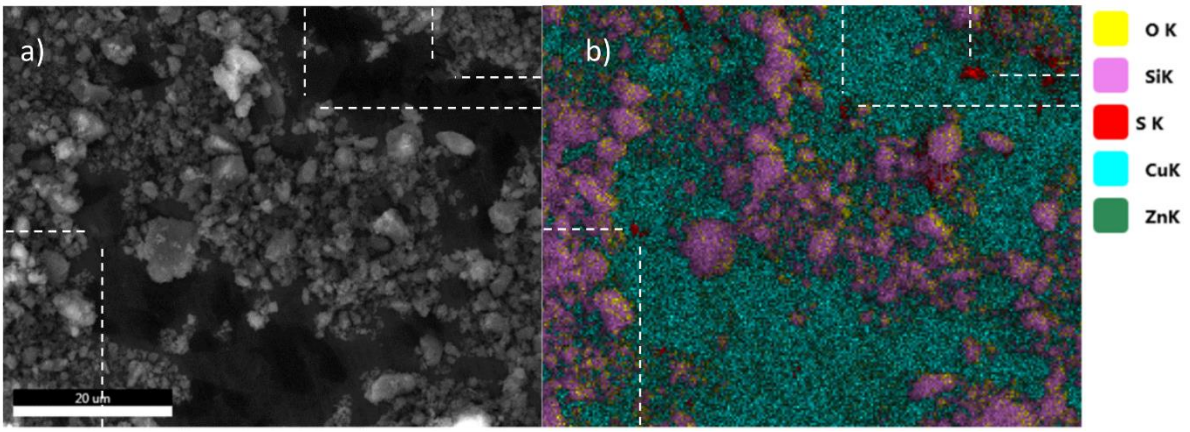

Figure S3: a) SEM image and b) EDX map of fused quartz. Dashed white lines indicate the position of EDX S signals on the SEM image. S is associated with the Cu / Zn substrate (S is also found on the bare substrate) rather than the fused quartz particles.

SEM shows that the fumed silica is indeed chain agglomerates similar in morphology to MSPs. This is in good agreement with previous work (Bogdan et al., 2003).

SEM shows that the fused quartz has a broad size distribution, with particles ranging from hundreds of nm to several µm in diameter. EDX detected no elements other than Si and O in these particles. Since the limit of detection here is on the order of 0.1 % this does not conclusively rule out contamination as a source of nucleation by the fused quartz material, however this is rarely practically possible.

Model Description

The Whole Atmosphere Community Climate Model (WACCM) is a 3D numerical high-top coupled chemistry-climate model extending vertically from the surface to about 140 km(Marsh et al., 2013), which is part of the NCAR Community Earth System model (CESM) (Hurrell et al., 2013). In this study, CESM v1.2.2 and WACCM 4 (CAM v5.3.57) were used. Aerosol properties are computed using CARMA (Community Aerosol and Radiation Model for Atmospheres) (Toon et al.,

1988;Toon et al., 1979;Turco et al., 1979), which employs a sectional (size bin) representation. Specified dynamics (SD) was used, with meteorological values nudged below 50 km using the Modern-Era Retrospective Analysis for Research and Applications (MERRA) reanalysis for an initial spin up period followed by conditions from July 2010 to July 2014 (Rienecker et al., 2011).

WACCM includes a detailed neutral chemistry for the middle atmosphere based on the Model for Ozone and Related Chemical Tracers (MOZART) (Kinnison et al., 2007) and sulfur chemistry(English et al., 2011); there are 66 species, including the sulfur-containing species OCS, S, SO, SO_2, SO_3, HSO_3, H_2SO_4, and a large set of photolysis, gas phase, and heterogeneous reactions (Lamarque et al., 2012). Emissions of SO_2, NO_2, NO, CO, NO, and DMS, and boundary conditions including for OCS were included (Lamarque et al., 2012). SO_2 emissions from both continuously and explosively erupting volcanos were also included(Mills et al., 2016). H_2SO_4 visible photolysis rates (Feierabend et al., 2006), including the pressure dependence (Miller et al., 2007), and Lyman α rates were also included(Lane and Kjaergaard, 2008). The aerosol surface area densities required to calculate the rates of heterogeneous reactions are obtained from CARMA. All simulations were performed on a 2.5° × 1.9° longitude/latitude grid with 88 model levels up to ~140 km, and CAM4 physics.

The CARMA aerosols here include two “groups”: pure sulfate and mixed sulfate, where mixed sulfate consists of sulfate with an MSP core. Both groups have 28 bins between 0.34 nm and 1.6 µm, with particle densities of 2.0 and 1.9 g cm^{-3} for pure sulfate and mixed sulfate, respectively. Pure sulfate is formed in the smallest size bin by homogeneous nucleation of H_2SO_4 and H_2O. MSPs are input into the smallest size bin of the mixed sulfate group (so that they begin as 100 % MSP core), between 75 and 110 km(Kalashnikova et al., 2000), with the input scaled to a selected global input rate from meteoric ablation (Carrillo-Sánchez et al., 2016).

Condensation and evaporation of H_2SO_4 occurs for both aerosol groups, but evaporation from mixed sulfate only occurs for particles that have a core fraction less than 0.5. For mixed sulfate, this condensation and limited evaporation mimics the reaction between H_2SO_4 and MSP metal atoms (Saunders et al., 2012). Coagulation occurs within and between both aerosol groups (English et al., 2012).

Output mass concentrations of cores (MSPs) for the 70 hpa level sampled at 67 °N (where Polar Stratospheric Clouds (PSCs) have been observed (Voigt et al., 2005)) is shown in Figure S4. The mean of February across these four years is used in the main body of the paper as an estimate of the available MSP mass.

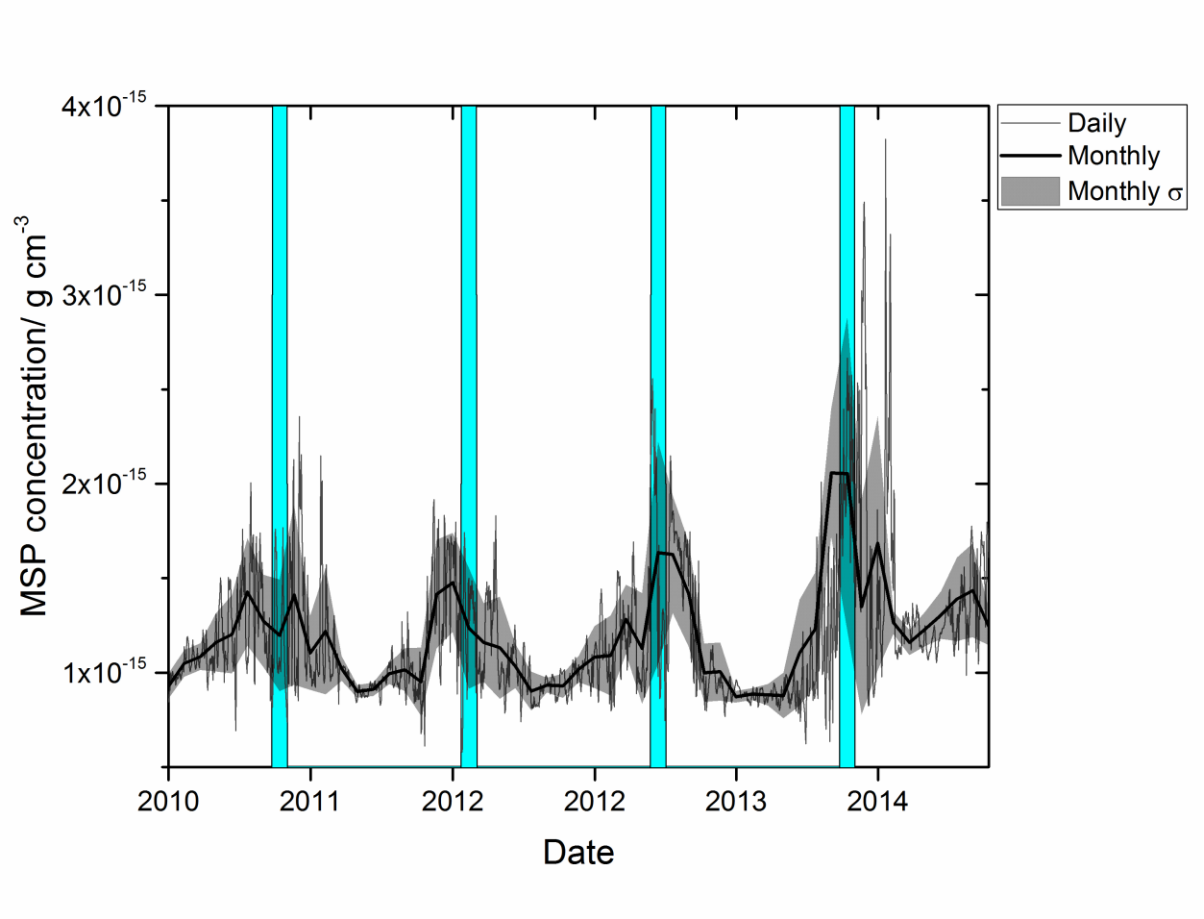

Figure S4: MSP mass concentrations from WACCM-CARMA. Zonally averaged data is shown for 67 °N and 70 hPa. The thin grey line shows daily mean data, thick black line monthly means, and the grey shaded area the monthly mean ± one standard deviation in the daily data. Vertical shaded areas indicate February in each year, when PSC are typically observed.

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