



The prevalence of meteoric-sulphuric particles within the stratospheric aerosol layer and their influence on how pure sulphuric particles are transported and transformed

Graham Mann (1,2), James Brooke (3), Kamalika Sengupta (1), Lauren Marshall (1), Sandip Dhomse (1), Wuhu Feng (1,2,3), Ryan Neely (1,2), Charles Bardeen (4), Nicolas Bellouin (5), Mohit Dalvi (6), Colin Johnson (6), Luke Abraham (7), Terry Deshler (8), Larry Thomason (9), and John Plane (3)

(1) School of Earth and Environment, University of Leeds, Leeds, United Kingdom, (2) National Centre for Atmospheric Science, University of Leeds, Leeds, United Kingdom, (3) School of Chemistry, University of Leeds, Leeds, United Kingdom, (4) Atmospheric Chemistry Observations and Modelling Laboratory, University of Colorado, Boulder, Colorado, USA, (5) Department of Meteorology, University of Reading, Reading, United Kingdom, (6) Hadley Centre for Climate Prediction and Change, UK Meteorological Office, Exeter, United Kingdom, (7) Department of Chemistry, University of Cambridge, Cambridge, United Kingdom, (8) Department of Atmospheric Science, University of Wyoming, Laramie, Wyoming, USA., (9) NASA Langley Research Center, Hampton, Virginia, USA

The widespread presence of meteoric smoke particles (MSPs) within a distinct class of stratospheric aerosol particles has become clear from in-situ measurements in the Arctic, Antarctic and at mid-latitudes. Knowledge of how extensive inclusions of MSPs occur within the liquid background stratospheric aerosol is important in light of growing evidence that many polar stratospheric clouds form heterogeneously in the Arctic.

In this study, we first explain the adaptation of the interactive stratosphere-troposphere aerosol-chemistry-climate model UM-UKCA to additionally resolve the heterogeneous nucleation of meteoric-sulphuric particles, alongside the existing pure aqueous sulphuric acid particles formed by homogeneous nucleation.

We present the simulated global distribution of both types of stratospheric aerosol particle showing how they form on 10-20nm MSP cores in the uppermost stratospheric aerosol layer at high latitudes, as MSPs are transported down from the mesosphere within the polar vortex. Our simulations suggest that meteoric-sulphuric particles contribute around 5-10 particles per cc additional condensation nuclei to the upper half of the winter polar stratospheric aerosol layer at particle sizes around 30-70nm. Although the meteoric-sulphuric particles are at lower concentrations in the mid-latitude stratosphere, resolving their presence in the model changes dramatically the simulated lifecycle of the background pure stratospheric sulphuric particles.

The additional condensation sink from the MSP-core particles reduces the condensational growth of the pure sulphuric particles, changing their vertical distribution, stratospheric residence time and transport trajectory within the prevailing Brewer-Dobson circulation. We find that resolving both sulphuric particle types in the model reduces the simulated stratospheric aerosol layer sulphur burden by a factor of 5 and greatly improves CN concentration predictions, remedying a previous high bias in the upper stratospheric aerosol layer from excessive particle formation in polar winter and spring.

Comparing to particle concentration measurements at Laramie, our results suggest that, aside from the observed layer of enhanced concentrations (at 30km) during late winter early spring (which is caused by homogeneous particle formation), in other months, the vertical profile of CN ($D_p > 10\text{nm}$) is approximately constant above 20km, the change from the decreasing profile from 15-20km caused both by the MSP-nucleated sulphuric particles and a strong additional seasonal variation (absent in previous simulations) caused by meridional transport of pure sulphuric particles homogeneously nucleated in the tropics.