Supplementary Text S2. Concentrations of reactants and phase partitioning

Concentrations of gas-phase and aqueous-phase reactants were estimated with a variety of procedures. For O3, SO2, and soot, estimates of atmospheric concentrations from a global simulation using the Mozart2 model were used (Horowitz et al., 2003; Ryaboshapko et al., 2007b). For OH•, global model results from Lu and Khalil (1991) were interpolated to create estimated concentrations dependent on latitude, elevation, month, and time of day. For total H2O2 and HCl concentrations, a constant, typical value equivalent to a gas phase mixing ratio of 1 ppb was used (Finlayson-Pitts and Pitts, 2000; Graedel and Keene, 1995). For total reactive chlorine, a constant value equivalent to a gas-phase mixing of 100 ppt was used for the lowest 100 m in the atmosphere over the ocean at night, following the approach of Bullock and Brehme (2002), consistent with the findings of Graedel and Keene (1995).

During each model time step within each grid cell, the liquid water content (LWC, g m-3) of the atmosphere was estimated based on the relative humidity (RH) using the following approach. For cells with RH less than a threshold RHcrit (80%) or with temperature less than 0 oC, LWC was assumed to be zero. For RH > RHcrit in all cells above the surface layer, a cloud fraction *fcloud* and associated LWC was estimated by



For surface layer cells, LWC was estimated following the methodology outlined by Fitzgerald (1975), assuming the particles were ammonium sulfate, starting with a dry particle size distribution as shown in Figure S1.The procedure essentially estimates the amount of water an ammonium sulfate particle of a given dry mass would have to absorb at a given relative humidity (*RH*) for the droplet’s water to be in vapor-liquid thermodynamic equilibrium. If the RH was below the deliquescence RH, the particles were assumed to be dry (i.e., LWC = 0).

If no liquid water existed in a given cell, only gas-phase reactions were utilized (reactions 1-5 in Table 1). If liquid water was present, then a more complex treatment was utilized. First, the gas-liquid and aqueous phase ionic equilibrium conditions were estimated, satisfying the relationships shown in Table S1, along with ionic and mass balances, using an iterative Newton-Raphson-based approach. In calculating these equilibrium conditions, a constant pH of 4.5 and a constant Cl-1 aqueous phase concentration of 2.5 mg/L was utilized in the simulation, following the approach of Ryaboshapko et al. (2007a). Then, the gas-phase and aqueous-phase reactions and transformations described in Table 1 were carried out.

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