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Solid-gas interactions in the eruption plume can both depress and enhance volcanic ash ice-nucleating activity

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Volcanic ash generated by explosive eruptions can act as ice-nucleating particles, promoting freezing of supercooled water droplets in the eruption plume and the ambient atmosphere, and so impacting processes such as plume electrification, ash aggregation, and cloud glaciation. Our initial study of a compositional range of milled ash and glass materials demonstrated that mineralogy is likely a key property influencing ice nucleation by ash.¹ However, the surface properties of ash are modified by interaction with magmatic gases in the hot core of the eruption plume, and it is not known how such in-plume interactions might affect the ice-nucleating activity (INA) of ash.

Here we investigated the influence of high temperature solid-gas interactions on the INA of three milled ash (Tungurahua, Astroni, Etna) and two milled mineral (K-feldspar, quartz) materials. Sub-samples of these materials were exposed to pure water vapour (H_2O) or mixtures of water vapour with $HCl_{(g)}$ (H_2O-HCl) or $SO_{2(g)}$ (H_2O-SO_2) under an 800 °C/400 °C heating sequence in the Advanced Gas-Ash Reactor.² The INA of the non-treated and treated samples was then assessed using a microlitre Nucleation by Immersed Particle Instrument.³ The H_2O treatment decreased the INA relative to that of the non-treated sample for all materials, and the H_2O-HCl treatment decreased the INA to the same extent or more. Conversely, the H_2O-SO_2 treatment increased the INA (Tungurahua ash, Etna ash), or decreased the INA 1) to a lesser extent than the other treatments (Astroni ash), 2) to the same extent as the other treatments (quartz), or 3) to a greater extent than the other treatments (K-feldspar).

The depression in INA induced in all cases by the H_2O treatment may relate to dehydroxylation of the silicate materials' surfaces at high temperatures. On the other hand, differing effects on INA of the H_2O-HCl and H_2O-SO_2 treatments is inferred to relate to contrasting reactivities of these materials towards $HCl_{(g)}$ and $SO_{2(g)}$. Water leachates of the samples suggest that chloride and sulphate salts (e.g., NaCl, CaSO₄) formed on the H_2O-HCl - and H_2O-SO_2 -treated ash surfaces, respectively, but not on the H_2O-HCl - and H_2O-SO_2 -treated mineral surfaces. Additional tests suggest that the changes in INA observed for these treated ash samples do not reflect a 'solute effect'⁴ imparted by the chloride or sulphate salts in water, implying that the ice-nucleating properties of the ash surfaces themselves are somehow changed by reaction with $HCl_{(g)}$ and $SO_{2(g)}$.

Surface-sensitive analyses could be useful to elucidate how sample surfaces have been modified

by the different solid-gas interactions at the scale relevant for ice nucleation, and so potentially shed light on the cause of the depression and enhancement in INA observed here. The possibility that in-plume reaction with $\text{SO}_{2(g)}$ can increase the INA of volcanic ash in particular merits further investigation, as a previous line of thought has been that exposure of silicate particles to this acidic gas decreases INA.

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³Whale et al. (2015) *Atmos. Meas. Tech.*, 8, 2437-2447. doi:10.5194/amt-8-2437-2015

⁴Whale et al. (2018) *Chem. Sci.*, 9, 4142-4151. doi:10.1039/C7SC05421A