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





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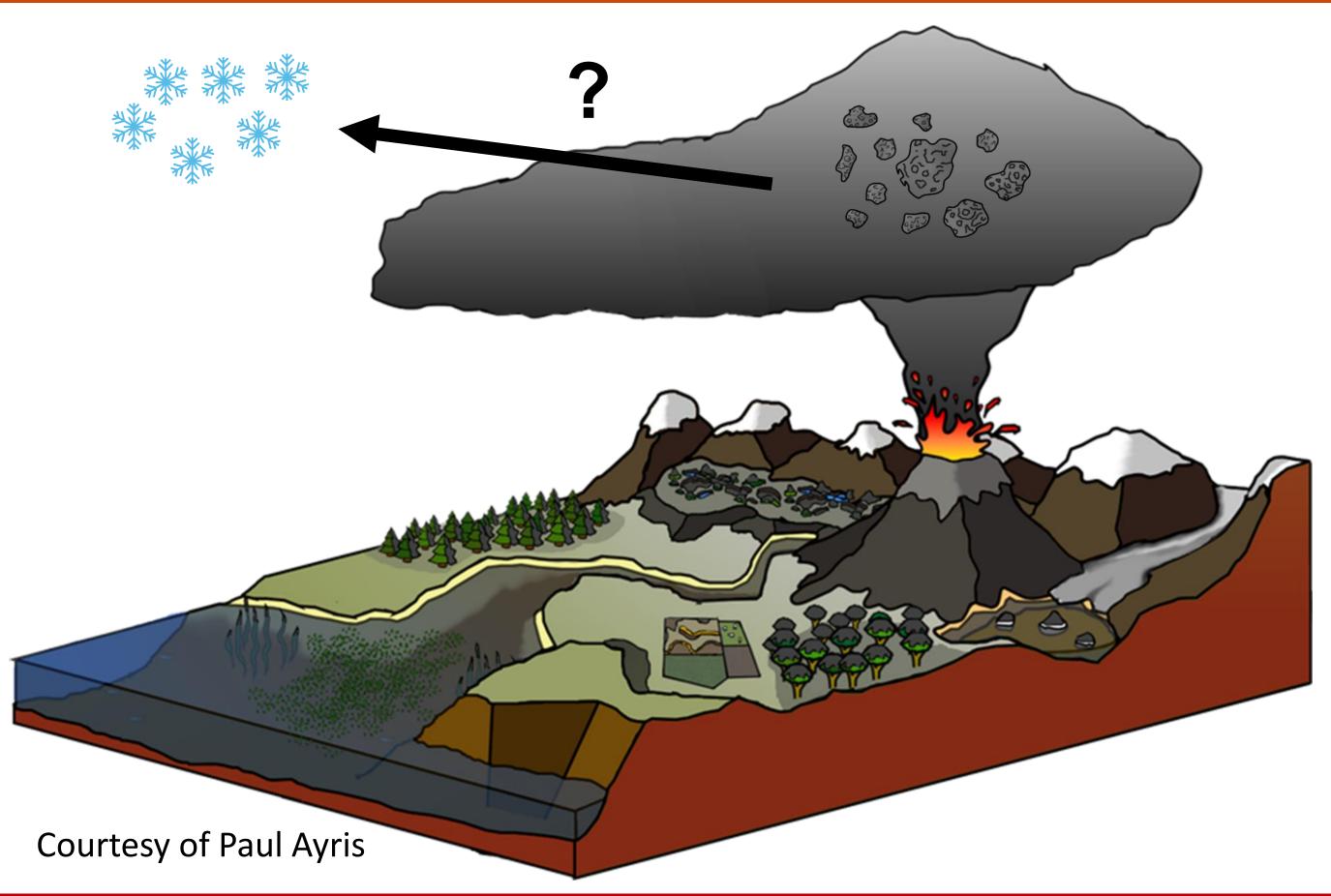
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Background

Volcanic ash can act as ice-nucleating particles, promoting freezing of supercooled water in the eruption plume and cloud and the atmosphere¹

It remains unclear what drives the large variation in ice-nucleating activity (INA) of ash reported in previous field and laboratory measurements

Our first study of a range of ash and glass samples suggests that crystalline phases (e.g., feldspar minerals) likely play an important role²

However, ash surface properties can be modified by interactions with magmatic gases in the hot eruption plume,³⁻⁵ and it is not known how such interactions might affect the INA of ash

Materials and Methods

Five silicate materials studied: Tungurahua tephra (TUN), Astroni tephra (AST), Etna tephra (ETN), K-feldspar (FELD), Smoky quartz (QRTZ)

Samples were exposed in an Advanced Gas-Ash Reactor⁶ at 800/400 °C to water vapour on its own (H₂O) or mixed with $SO_{2(g)}$ (H₂O-SO₂) or $HCl_{(g)}$ (H₂O-HCl) for 1200 seconds



Figure 1. Photo of the Advanced Gas-Ash Reactor showing the working tube within a three-stage furnace

Soluble compounds formed on samples were assessed by leaching in water, filtration and analysis by ICP-OES and IC

Ice-nucleating activity of samples was assessed by cold stage droplet assays using a microlitre Nucleation by Immersed Particles Instrument⁷

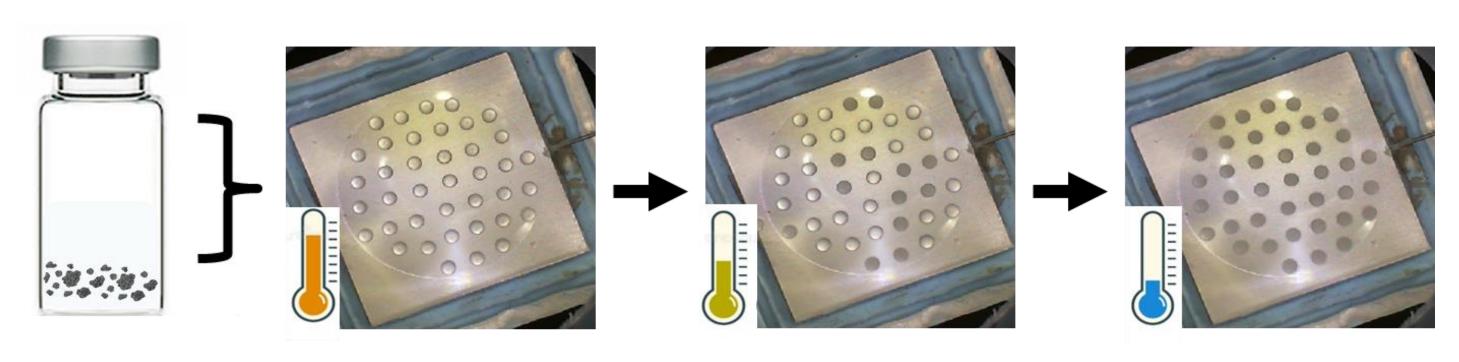


Figure 2. Schematic of a droplet assay experiment showing water droplets containing 1 wt.% sample suspension freezing as temperature decreases

Ice-nucleation active site density per unit solid surface area (n_{c}) as a function of temperature was calculated as:

$$n_{ice}(T)/n = 1-\exp(-n_s(T)A)$$

 $n_{ice}(T)$ = number of droplets frozen at temp (T)

n = total number of droplets

A = total solid surface area per droplet

Results Summary Figure

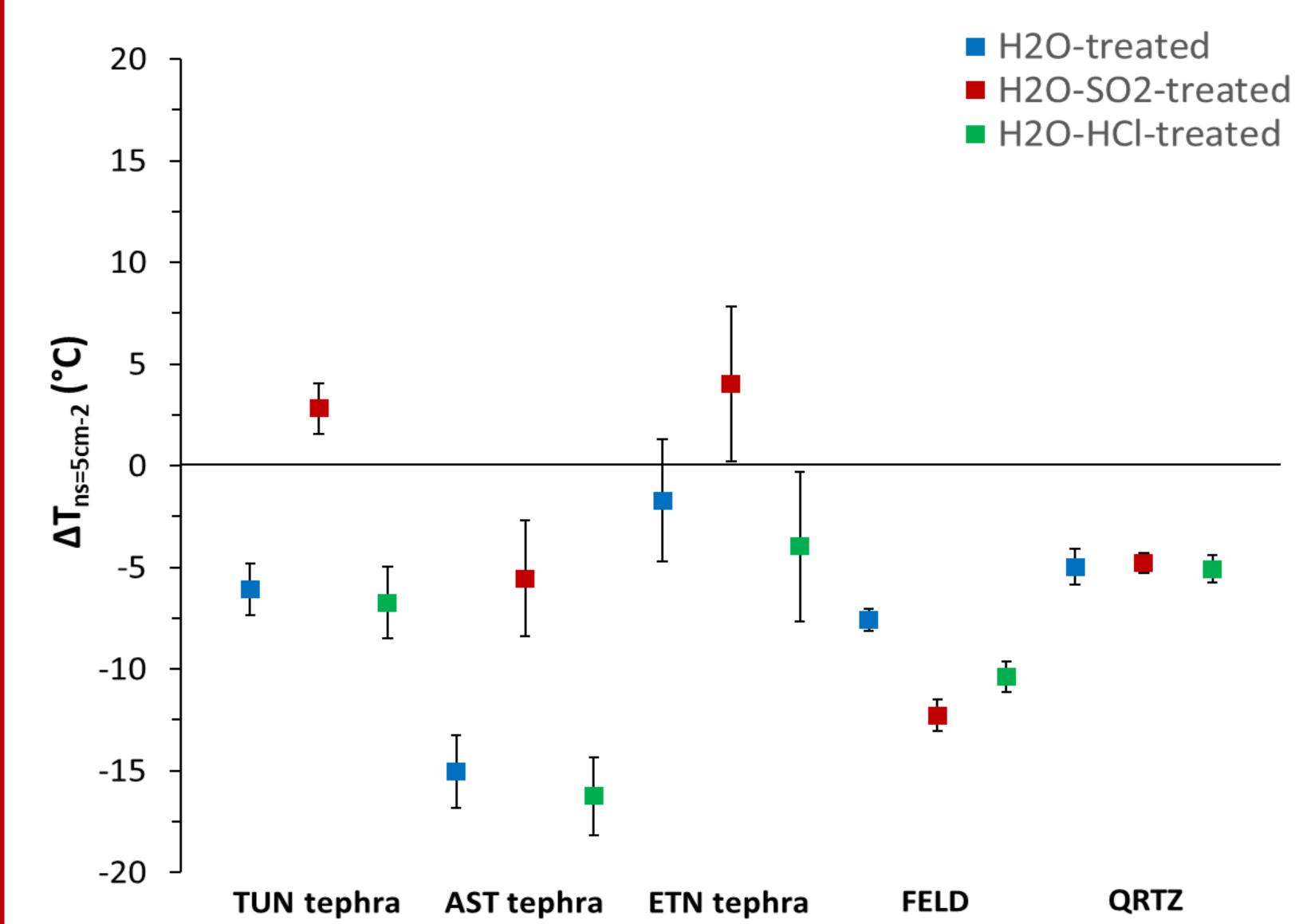


Figure 3. Difference in INA ($\Delta T_{ns \approx 5 \text{ cm}-2}$) between the H₂O-treated, H₂O-SO₂-treated, or H₂O-HCl-treated and the non-treated silicate materials. Values above or below the horizontal black line indicate that the INA of the treated samples is higher or lower, respectively, than the INA of the non-treated samples. The error bars reflect the ranges of values based on the standard deviations of mean $T_{ns \approx 5 \text{ cm-}2}$ values for the treated and non-treated samples from replicate ice nucleation experiments.

H₂O and H₂O-HCl decreased INA relative to the non-treated material

H₂O-SO₂ either *increased* INA relative to the non-treated material (TUN and ETN) or decreased INA to a lesser extent (AST), to a greater extent (FELD), or to the same extent (QRTZ) as the other treatments

Differing effects of treatments likely reflect contrasting reactivities of the silicate materials towards H_2O and $SO_{2(g)}$ or $HCl_{(g)}$

Changes in tephra INA do not relate to a 'solute effect'8,9 of sulphate or chloride salts formed by reaction with $SO_{2(g)}$ or $HCl_{(g)}$ at high temperatures

¹Durant et al. (2008) *J Geophys Res*, 113, D09206 ²Maters et al. (2019) *Atm Chem Phys*, 19, 5451-5465 ³Delmelle et al. (2007) *Earth Planet Sci Lett*, 259, 159-170 ⁴Maters et al. (2016) *Earth Planet Sci Lett*, 450, 254-262 ⁵Delmelle et al. (2018) *Rev Mineral Geochem*, 84, 285-308 ⁶Ayris et al. (2015) *Bull Volcanol*, 77, 104 ⁷Whale et al. (2015) *Atm Meas Tech*, 8, 1-11 ⁸Whale et al. (2018) *Chem Sci*, 9, 4142-4151 ⁹Kumar et al. (2019) *Atm Chem Phys,* 19, 6059–6084