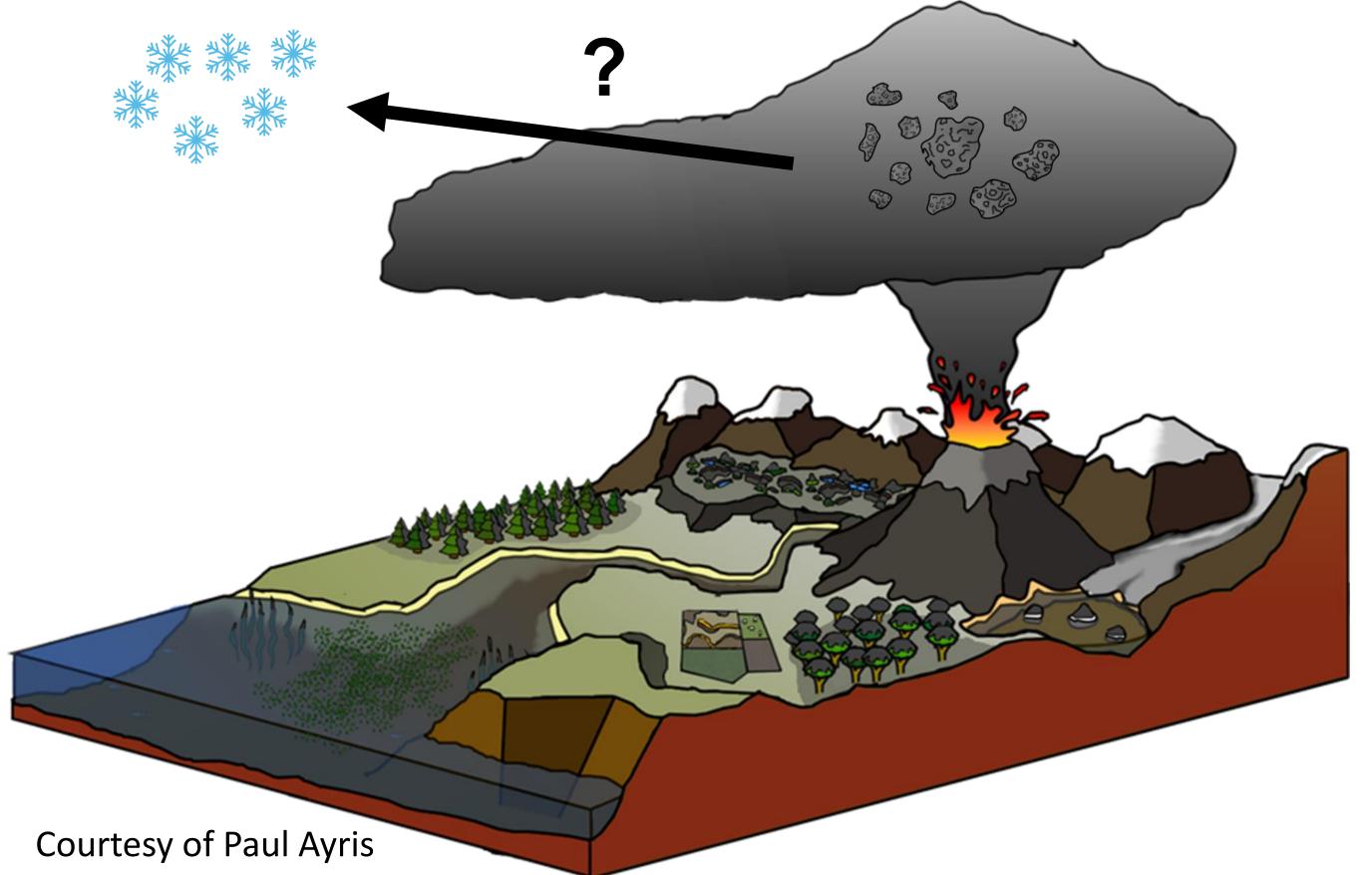
Solid-gas interactions in the eruption plume can both depress and enhance volcanic ash ice-nucleating activity

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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 atmosphere<sup>1</sup>

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 role<sup>2</sup>

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

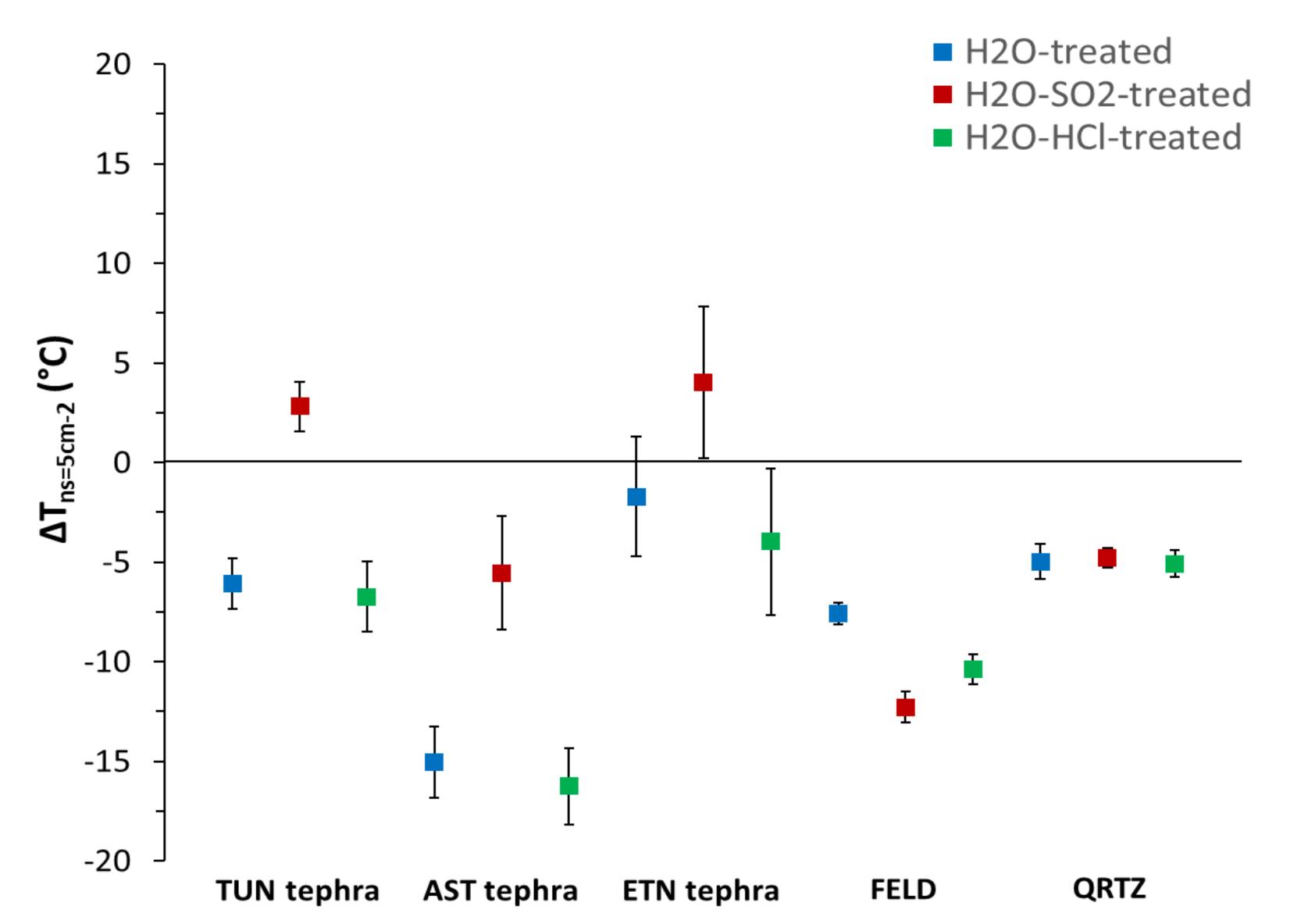
## **Materials and Methods**

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 Reactor<sup>6</sup> at 800/400 °C to water vapour on its own ( $H_2O$ ) or mixed with  $SO_{2(g)}$  ( $H_2O$ -SO<sub>2</sub>) or  $HCl_{(g)}$  ( $H_2O$ -HCl) for 1200 seconds



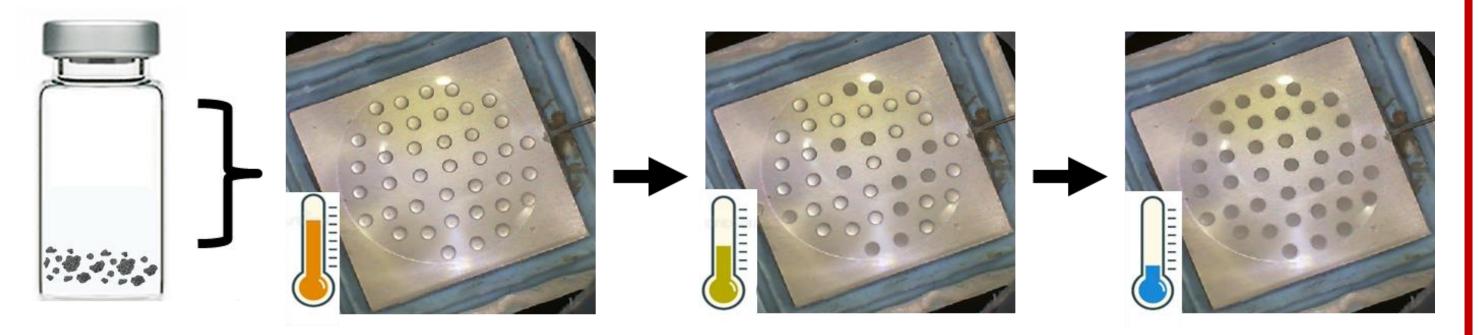
## **Results Summary Figure**



**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 Instrument<sup>7</sup>



**Figure 3.** Difference in INA ( $\Delta T_{ns \approx 5 \text{ cm}-2}$ ) between the H<sub>2</sub>O-treated, H<sub>2</sub>O-SO<sub>2</sub>-treated, or H<sub>2</sub>O-HCI-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<sub>ns ≈ 5 cm-2</sub> values for the treated and non-treated samples from replicate ice nucleation experiments.

H<sub>2</sub>O and H<sub>2</sub>O-HCl *decreased* INA relative to the non-treated material

 $H_2O-SO_2$  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

**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  $(n_s)$  as a function of temperature was calculated as:

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

- $n_{ice}(T)$  = number of droplets frozen at temp (T) n = total number of droplets
- A = total solid surface area per droplet

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'<sup>8,9</sup> of sulphate or chloride salts formed by reaction with SO<sub>2(g)</sub> or HCl<sub>(g)</sub> at high temperatures

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