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Hot volcanic ash filters eruptive SO2 during hot transport in conduits and the lower plume: A predictive model with implications for the climate impacts of volcanic eruptions

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Sulfate aerosols are a primary driver of climate impacts during and following volcanic eruptions and form from erupted SO_2 gas. However, the amount of SO_2 that is delivered to the stratosphere is not clearly related to the amount dissolved in the magma (the 'sulfur excess problem'). Therefore, magma properties and eruption magnitude are not necessarily predictive of climate impacts from eruptions, which is exacerbated by the as-yet unknown importance of the insulated, hot transport pathway. During a magnitude 6 explosive volcanic eruption there is up to 100 seconds of transport between the magma fragmentation depth – where volcanic ash is formed and the mixture accelerates – and the Earth's surface. Here, we present a numerical implementation of a theoretical framework which predicts the rapid reactions between gases and volcanic ash in this transport interval, which include: (1) iron oxidation state changes; (2) SO_2 uptake via calcium sulfate surface crystallization; (3) HCl uptake via NaCl surface crystallization; and (4) incipient nanolite crystallization that may be related to (1). In all cases, these processes are rate-limited by a suite of diffusive exchanges between the ash bulk and surface, for which our model solves. To demonstrate the upscaled importance of these processes, we couple our models to volcanic plume simulations (using a 1991 Pinatubo baseline simulation), and output the bulk SO_2 that can be captured by ash. We find that depending on the source parameters of the eruption, anywhere between 30 and 100 wt.% of the total erupted SO_2 can be removed from the plume gas and captured by ash. This effectively changes the sink of SO_2 from the stratosphere to the hydrosphere, as $CaSO_4$ crystals are soluble and ultimately wash into the environment following ash deposition. We propose that these hot sulfur scrubbing processes may be crucial in mediating SO_2 delivery to the atmosphere, and therefore may explain much of the complexities associated with correlating eruption magnitude with climate impacts in the recent past or back into the Last Glacial period.