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Sulfur solubility and oxidation state in lower crustal arc dacitic magmas: implications for the 1991 Mt. Pinatubo eruption

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The role of sulfur (S) in silicate melts is crucial in controlling crustal processes related to ore formation or volcanic outgassing, yet the complexities of sulfur behavior in magma remain incompletely understood. To investigate the dependence of the transition of sulfur oxidation state on melt composition and pressure in arc magmas at lower crustal levels, we performed piston-cylinder experiments on sulfur-saturated, hydrous dacitic melt at 700 MPa and 950 °C using various fO_2 solid-state buffers (fO_2 ranging from FMQ -2 to FMQ +4.5; FMQ = fayalite-magnetite-quartz oxygen buffer). As starting glass compositions, we chose synthetic, anhydrous dacitic glasses corresponding to Pinatubo dacite in compositions. To address the compositional effect, two glass compositions with (1) Fe-bearing and (2) Fe-free synthetic glasses were prepared. The new experimental data reveal that the amount of sulfur dissolved in the dacitic melt range from 68 ± 11 to 1328 ± 20 ppm with increasing fO_2 and there is no significant difference in sulfur solubility between the iron-bearing and iron-free melts. Our results indicate the sulfide-sulfate transition occurs between FMQ+1 and +2 in dacite (at 950-1000 °C, 300-700 MPa) with a negligible pressure effect. Compared to previous work on basaltic melt, sulfide-sulfate transition in dacitic melt is shifted to higher fO_2 (by +0.4 log unit). Our experimental data suggest that the parental magma from Pinatubo was oxidized with fO_2 > FMQ +0.45. However, the S saturation contents found in experiments cannot explain the high sulfur contents measured in some of the Pinatubo dacite melt inclusions, indicating additional supply of sulfate (e.g. by hydrothermal fluids). We further investigated the degassing trends of key volatile species using the volatile degassing models D-Compress and Sulfur_X. These models consider the complexity of S concentrations and speciation across varying melt and fluid compositions, pressures, temperatures, and fO_2. Our results show that even with the increased solubility at higher pressures and oxidation states, the recorded S in melt inclusions does not fully explain the observed substantial SO_2 emissions, implying the existence of an additional S^{6+} source.