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Revisiting the gas-phase rate coefficient of the OH + SO_2 + M → HSO_3 + M reaction

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Oxidation of SO_2 to sulfuric acid impacts acid precipitation and aerosol nucleation in Earth's atmosphere in remote and polluted environments. This oxidation can take place in both the liquid and gas phase. Only the gas phase oxidation is expected to lead to new particles because of the clustering reactions of H_2SO_4. This aerosol nucleation has a major effect upon air quality and the Earth's radiative balance, and is of crucial importance to the chemistry of the atmosphere.

The rate limiting step in this process is the reaction of OH radicals with SO_2 to form HSO_3. The pressure- and temperature-dependent reaction of OH + SO_2 has been studied many times previously – since its importance was first recognized in the 1970s. Notwithstanding, some of the most recent literature has cast doubt on much of this data, especially under conditions that are relevant to atmospheric chemistry.

Here, we present measurements of the rate coefficient using the pulsed laser photolysis–laser induced fluorescence technique as a function of temperature (249–373 K) and of pressure in helium, argon and nitrogen bath gases (30–600 Torr). In addition, relative rate measurements using a chamber at 760 Torr (N_2, O_2 and air) were also performed to corroborate our absolute observations. By utilizing these new data, together with the available literature data, an updated pressure- and temperature-dependent parameterization will be provided. This allows the atmospheric impact of this reaction to be constrained with a new level of certainty.

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