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An isotopic view of ionising radiation as a source of sulphuric acid

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Sulphuric acid is an important factor in aerosol nucleation and growth. It has been shown that ions enhance the formation of sulphuric acid aerosols, but the exact mechanism remains undetermined. Furthermore some studies have found a deficiency in the sulphuric acid budget, suggesting a missing source. In this study the production of sulphuric acid from SO_2 through a number of different pathways is investigated. The production methods are standard gas phase oxidation by OH radicals produced by ozone photolysis with UV light, liquid phase oxidation by ozone, and gas phase oxidation initiated by gamma rays. The distributions of stable sulphur isotopes in the products and substrate were measured using isotope ratio mass spectrometry. All methods produced sulphate enriched in 34S and we find a d34S value of 8.7 $\pm$ 0.4 ‰ (1 standard deviation) for the UV-initiated OH reaction. Only UV light (Hg emission at 253.65 nm) produced a clear non-mass-dependent excess of 33S. The pattern of isotopic enrichment produced by gamma rays is similar, but not equal, to that produced by aqueous oxidation of SO_2 by ozone. This, combined with the relative yields of the experiments, suggests a mechanism in which ionising radiation may lead to hydrated ion clusters that serve as nanoreactors for S(IV) to S(VI) conversion.