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High Resolution Photoabsorption Cross-Sections of Isotopologues of SO_2

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The timing of the oxygenation of the Earth's atmosphere is a central issue in understanding the Earth's paleoclimate. The discovery of mass-independent fractionation (MIF) of sulphur isotopes deposited within Archean and Paleoproterozoic rock samples has given rise to a possible marker, through the transition between MIF within older rock samples (> 2.4 Gyrs) to mass-dependent fractionation (MDF) within younger samples, for the rise in oxygen concentrations within the Earth's atmosphere [Farquhar,2003].

Laboratory experiments [Farquhar,2001][Pen,2008] suggest isotopic self shielding during the gas phase photolysis as the dominant mechanism for MIF. Self shielding is present for SO_2 at wavelengths shorter than 220 nm where it undergoes partial predissocation. The UV absorption of SO_2 is dominated by the $\tilde{C}^1B_2$-$\tilde{X}^1A_1$ electronic system which comprises of strong vibrational bands extending from 170 - 230 nm. Within an atmosphere consisting of low O_2 and O_3 concentrations, such as that predicted for the early Earth, UV radiation would penetrate deep into the ancient Earth's atmosphere within the 180 - 220 nm range driving the photolysis of SO_2.

We have conducted the first ever high resolution measurements of the photo absorption cross sections of several isotopologues of SO_2, namely $^{32}SO_2$, $^{33}SO_2$, $^{34}SO_2$ and $_{36}SO_2$. The cross sections are being measured at Imperial College at initial resolutions of 1.0 cm^{-1} which will be increased to resolutions < 0.5 cm^{-1} for inclusion in photochemical models of the early Earth's atmosphere. The models will be used to more reliably interpret the sulphur isotope ratios found within ancient rock samples [Lyons, 2007].

Initial 1.0 cm^{-1} resolution measurements of several isotopologues of SO_2 will be presented in addition to preliminary < 0.5 cm^{-1} photo absorption cross section measurements.