Medium and high-resolution UV cross sections for isotopic SO_2

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The prevailing interpretation of the Archean sulfur mass-independent fractionation (S-MIF) signature has been that photodissociation of SO_2 in a low O_2 atmosphere allows for the formation of multiple sulfur reservoirs with different S-MIF signatures, which then become recorded in barites and pyrites in sediments. The key assumption has been that SO_2 dissociation yields a S-MIF signature. We are measuring cross sections for isotopic SO_2 to both understand the formation of any S-MIF signature produced during SO_2 photolysis, and to use in photochemical models of the early Earth atmosphere.

We used the VUV FTS of the DESIRS beamline at the Soleil synchrotron to measure cross sections. The high signal to noise ratio, and rapid scanning capability of the VUV FTS allowed us to complete an extensive set of absorption measurements at 0.5 cm^{-1}, along with a first set of high resolution absorption measurements at ~ 0.1 cm^{-1}. We focused on the ~190-220 nm dissociation region of SO_2. The 0.5 cm^{-1} data is used to evaluate S-MIF signatures from SO_2 photolysis, including those due to any intensity variations, while the 0.1 cm^{-1} is to be used primarily for line positions and the development of rovibronic models for C-X band system in isotopic SO_2. We also made quadrupole mass spectrometry measurements on each of the gas bottles to test for impurities. A 2.5% mass 44 impurity (CO_2) was found in our $^{32}SO_2$ bottle, which must be accounted for in order to obtain accurate cross sections.

Integrated 0.5 cm^{-1} cross sections over 190-220 nm yield values for each of the isotopes that are within < 1% of each other. This means that it is unlikely that optically-thin photolysis of SO_2 is solely responsible for the S-MIF signature in ancient rocks.