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Isotope effect in the visible light photolysis of O_3 and implications for the isotope effect in the O_3 formation reaction

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The unusual isotopic composition of atmospheric O_3 has been primarily attributed to a strong mass independent isotope effect in its formation reaction. We show that visible light photolysis contributes significantly to the isotope enrichment of O_3 both in laboratory photolytic recycling experiments and throughout most of the troposphere. The fractionation due to photolysis is mass dependent. We also determined the (low-resolution) wavelength dependence of the fractionation associated with photolysis and find a much weaker wavelength dependence compared to semi-empirical model calculations. The fractionation in photolysis is shown not to depend on temperature, and the temperature dependent isotope enrichments found in photolysis recycling experiments can be corrected for the effect of photolysis to yield for the first time isotope effects in the O_3 formation reaction only. These enrichments are then used to reassess the relative rate coefficients of several O_3 – forming isotopic reaction channels.