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# Statistical clumped isotope signatures

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High precision measurements of molecules containing more than one heavy isotope in environmental samples are becoming available with new instrumentation and may provide novel constraints on element cycles in nature. These so-called clumped isotope signatures are reported relative to the random (stochastic) distribution of heavy isotopes over all available isotopocules of a molecule, which is the conventional reference. When multiple indistinguishable atoms of the same element are present in a molecule, this reference is calculated from the bulk isotopic composition of the molecule, which for rare heavy isotopes is approximated by the arithmetic average of the isotope ratios of single substituted atoms. We show here that this referencing convention leads to apparent negative clumped isotope anomalies when the indistinguishable atoms are from isotopically different populations. Such statistical clumped isotope anomalies must occur in any system where two or more indistinguishable atoms of the same element, but with different isotopic composition, combine in a molecule and these anomalies have to be taken into account in data interpretation. The size of the signal is closely related to the relative standard deviation of the initial isotope ratios of the indistinguishable atoms that have combined. Therefore, a measured statistical clumped isotope anomaly may allow assessment of the heterogeneity of the isotopic pools of atoms that are the substrate for formation of molecules.