Geophysical Research Abstracts
Vol. 15, EGU2013-9440, 2013
EGU General Assembly 2013

Inferring marine sinks and sources of monohalomethanes from their carbon stable isotope composition

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Within the last years much progress in quantifying the global emissions of various halocarbons has been made. However, the current emission estimates are still assigned with large uncertainties due to the inevitably large spatial and temporal variability in observed halocarbon mixing ratios and fluxes. An improved understanding of the biogeochemical controls of the production – destruction equilibrium may substantially reduce these uncertainties and is of vital importance to address potential future changes. The δ13C values of monohalomethanes vary sensitively towards changes of both, sources and sinks making them a valuable tool to assess concurrent production and degradation processes. Here we report carbon isotope ratios for chloromethane (CH3Cl) and bromomethane (CH3Br) in background air and coastal and open ocean surface waters. The samples were taken during five sampling campaigns between September 2010 and July 2012 with the sample locations spanning from 10°N to 60°N

Our results show an enrichment by about 4‰ for chloromethane in marine air masses (-36‰ as compared to continental air masses (-40‰ corroborating earlier findings. This enrichment is supported by the observation of even more enriched chloromethane in the ocean surface waters averaging -28‰ for the subtropical North East Atlantic. For bromomethane, our data show an even more pronounced enrichment by 16‰ from -44‰ in continental air masses to -28‰ in marine air masses. These isotopic differences can be attributed to the air sea exchange of these compounds in concert with the production – decomposition dynamics in surface oceans. Hydrolysis, assigned with an ε of 45‰ is regarded as the most important degradation process for chloromethane in surface oceans. Bromomethane from both, intrinsic sources and from the atmosphere, is known to be rapidly degraded in marine surface waters by biotic and abiotic processes. The abiotic degradation due to hydrolysis and transhalogenation is assigned with a large ε of 69 ± 8‰ $andthedegradationofbromomethanebymethylotrophicbacteriaisassignedwithan\varepsilon$ of 45 ± 10. First simple model studies indicate that these degradation mechanisms may be sufficient to explain the observed isotopic shift for bromomethane but that hydrolysis alone cannot account for the isotopic shift observed for chloromethane. These results provide new information for further elucidating the oceanic controls of the global chloromethane and bromomethane cycles.