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New insight into the atmospheric chloromethane budget gained using stable carbon isotope ratios

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Atmospheric chloromethane (CH_3Cl) plays an important role in stratospheric ozone destruction, but still many uncertainties exist regarding strengths of both sources and sinks and the processes leading to formation of this naturally occurring gas. The sinks of atmospheric CH_3Cl are now well established whereas in contrast no such concensus can be reached for the sources. Previously, we identified a new source of CH_3Cl, which can explain its formation in a variety of terrestrial environments (Hamilton et al. 2003). We demonstrated that the widespread structural plant component, pectin, reacts readily with chloride ion to form CH_3Cl at both ambient and elevated temperatures. We proposed that this abiotic chloride methylation process in terrestrial environments could be responsible for formation of a large proportion of atmospheric CH_3Cl. However, more information is required to determine the global importance of this new source and its contribution to the atmospheric CH_3Cl budget.

A potentially powerful tool in modelling the atmospheric CH_3Cl budget is the use of stable carbon isotope ratios. Very recently it has been reported that the reaction of CH_3Cl with OH radical, the dominant sink for atmospheric CH_3Cl, is accompanied by an unexpectedly large fractionation factor (C. Nielsen, personal communication). Also

recently we reported that CH_3Cl formed by the abiotic methylation process at ambient temperatures has a unique stable carbon isotope signature, extremely depleted in ^{13}C, unequivocally distinguishing it from all other known sources (Keppler et al. 2004). By taking into account these very recent findings and using existing literature data we present three scenarios for an isotopic mass balance for atmospheric CH_3Cl. Our calculations strengthen the idea that the bulk fraction of atmospheric CH_3Cl (1.7 to 2.6 Tg) is produced by an abiotic chloride methylation process in terrestrial ecosystems, primarily located in tropical and subtropical areas, where turnover of biomass is highest. Furthermore our findings also show that the microbial CH_3Cl sink is much larger than that previously assumed.

References

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