A cryogen-free automated measurement system of stable carbon isotope ratio of atmospheric methane

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Methane is an important greenhouse gas emitted from both natural and anthropogenic sources. Accurate understanding of various methane sources is needed for effective reduction of its emissions. Measurement of stable carbon isotope ratio of atmospheric methane is useful for top-down evaluation of relative contributions of various types of methane sources, because individual methane sources have characteristic isotope signatures. Isotope ratio data for atmospheric methane therefore receive increasing attention for constraining the global methane budget. To meet research community demand, it is imperative for measurement systems to achieve adequate precision and traceability as well as high measurement throughput to process number of air samples that are routinely shipped from monitoring sites to laboratory. Stable carbon isotope ratio has been measured by continuous-flow isotope ratio mass spectrometry where methane in sample air is preconcentrated in a trap filled with an adsorbent cooled by liquid nitrogen. By using a stirring cooler instead, we set up a cryogen-free measurement system, and our configuration attained reproducibility of ~0.1 per mil. This precision is comparable to those previously achieved by using liquid nitrogen; our system is however advantageous for unattended and automated measurement operation. We examined measurements for air sampled outside our laboratory, and the preliminary measurement data at ~1-hour intervals show a clear diurnal variation.

Keywords: Methane, Stable carbon isotope ratio

