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Atmospheric Methane 14C Analysis at the HEKAL AMS Facility

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The most powerful anthropogenically influenced greenhouse gas after carbon dioxide is methane. Its atmospheric mixing ratio is much lower than of CO2, but the global warming potential of CH4 is more than eighty times higher, on a 20-year timescale (IPCC, 2021). The atmospheric amount of CH4 has been growing rapidly since 2007, and the reasons are unclear. Several types of sources can contribute to this increase, but there is no scientific consensus on the contributions of the possible sources. Recently the global atmospheric 13C/12C (d13C) ratio in CH4 is decreasing, but d13C measurements of CH4 alone still cannot provide sufficient information about emission sources.

That is why atmospheric CH4 studies are frequently coupled with 14C isotope measurements. Measurements of radiocarbon (14C) in methane (CH4) may provide a method for identifying regional CH4 emissions from fossil versus biogenic sources because adding 14C-free fossil carbon reduces the 14C/C ratio (Δ14CH4) in atmospheric CH4 much more than biogenic carbon does.

In collaboration with the IMAU group at UU we have set up a new system for ambient air methane sample preparation for 14C analyses. Our aim was to reduce the necessary sample size below 30 dm3, to make sampling and transport easier. As methane amount in ambient air is about 200 times less (around 2 ppm) in ambient air than CO2, it is very important to eliminate the CO2 content. For this purpose, we have modified a bit the CH4 extraction line at IMAU Lab, to fit for 20-30 dm3 air samples, instead of the 1000-2000 dm3 size, which they normally use for stable and clumped isotope analyses of atmospheric CH4. After several cryogenic focusing steps, the air sample is rather enriched in CH4, and finally a gas chromatograph is used for complete separation from CO2 and CO. The cleaned CH4 gas samples were then transported and combusted in sealed tubes at HEKAL laboratory, and the cleaned CO2 gas product was measured by our LEA type AMS system in our Lab using its gas ion source interface (0.005-0.050 mg C). In this study we present the sample recovery, the preparation blank level, and the analytical uncertainty connected to the whole, complex sample preparation and 14C analyses method. A few examples will also be presented where real atmospheric samples (from elevated sources, city, and background regions) were measured.

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Student Submission

Yes

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