Radiocarbon measurements of atmospheric methane

Sönke Szidat[1], Christophe Espic[1]

[1]Department of Chemistry and Biochemistry & Oeschger Centre for Climate Change Research, University of Bern, Bern, Switzerland

szidat@dcb.unibe.ch

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1. Project description

Methane (CH_4) contributes substantially to global warming as the second most important anthropogenic greenhouse gas. Despite all efforts, individual sources of CH_4 remain poorly quantified and not well understood, and climate change could dramatically increase CH_4 natural emissions (Dlugokencky et al., 2011). Radiocarbon (^{14}C) measurements of atmospheric CH_4 can be used to evaluate the proportion of fossil sources (e.g. natural gas, fossil-fuel combustion) and contemporary sources (e.g. agriculture, wetlands) (Lassey et al., 2007). We developed a new CH_4 preconcentration and purification setup, which allows $^{14}CH_4$ measurements of atmospheric air (Espic et al., 2019). We currently investigate three strategic sites in Switzerland: the Beromünster tall tower (rural area), the Jungfraujoch research station (continental background) and the Department of Chemistry and Biochemistry in Bern (urban area). The comparison of the results from Beromünster and Jungfraujoch is of special importance, as it provides the potential of quantifying contributions of fossil and contemporary sources as well as $^{14}CH_4$ emissions of nuclear power plants for the rural site. Such a comparison has been performed in a similar way for the quantification of emissions of carbon dioxide (CO_2) using radiocarbon measurements (i.e. of $^{14}CO_2$) since 2012 (Berhanu et al., 2017).

Figure 1. Collection of air samples at Jungfraujoch. Picture: Ruedi Käser.

For a proof-of-principle study, in total 23 air samples were collected every second week at Jungfraujoch from January to October 2019. During one hour, ~100 L (at STP) of ambient air from the Sphinx were filled into PE-AL-PE bags (TESSERAUX, Germany) (Fig. 1) and transferred to Bern. Moreover, one overnight sampling was performed on 20th/21st March with a time resolution of 3 hours. In Bern, CH_4 and CO_2 were extracted from the air samples using a preconcentration and purification setup that involves a gas-chromatography (GC) separation of the carbon-containing gases (Fig. 2). The ^{14}C content of both gases was measured at the Laboratory for the Analysis of Radiocarbon with AMS (LARA) at the University of Bern (Szidat et al., 2014).

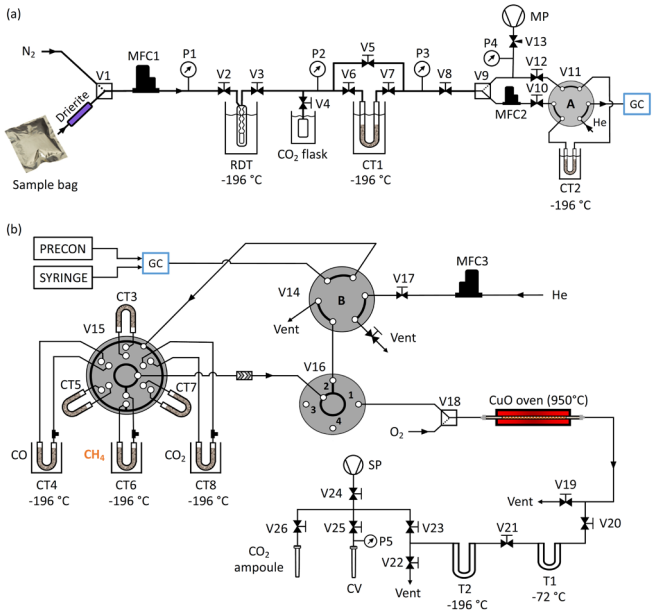

Figure 2. The setup for the preparation of atmospheric methane samples for radiocarbon analysis at the University of Bern (Espic et al., 2019), consisting of (a) the methane preconcentration setup (PRECON) and (b) the methane purification setup.

The study from 2019 indicated that Jungfraujoch is well suited as continental background station for $^{14}CH_4$. The variability of the individual measurements from Jungfraujoch was much smaller than for the rural and the urban sites. The continuation of our measurements will provide an update of $^{14}CH_4$ from a continental background site, which has not been determined since the early 2000s, when such a measurement was reported for Baring Head, New Zealand (Lassey et al., 2007). This data will serve as reference for regional source apportionment of methane using ^{14}C, i.e. by comparison with sites that are affected by local or regional methane emissions.

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Collaborating partners / networks

Prof. M. Leuenberger, Climate and Environmental Physics, University of Bern
Dr. D. Brunner, Dr. S. Henne, Empa, Dübendorf
Dr. S. Hammer, Institute of Environmental Physics, Heidelberg University

Address

Laboratory for the Analysis of Radiocarbon with AMS
Department of Chemistry and Biochemistry
University of Bern
CH-3012 Bern
Switzerland

Contacts

Prof. Dr. Sönke Szidat
Tel.: +41 31 631 4308
e-mail: szidat@dcb.unibe.ch