

Radiocarbon measurements of atmospheric methane

Thomas Laemmel¹, Michael Staub¹, Sönke Szidat¹

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

soenke.szidat@unibe.ch, thomas.laemmel@unibe.ch

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1. Project description

Methane (CH₄) contributes substantially to global warming as the second most important anthropogenic greenhouse gas. Despite all efforts, individual sources of CH₄ remain poorly quantified and not well understood, and climate change could dramatically increase CH₄ natural emissions (Dlugokencky et al., 2011). Radiocarbon (¹⁴C) measurements of atmospheric CH₄ 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). This approach benefits from the fact that ¹⁴C is extinct in fossil fuels due to their old age, whereas contemporary sources show a modern radiocarbon level (Szidat, 2020).

While ¹⁴CO₂ measurements are today an established method (Berhanu et al., 2017, Levin et al., 2003), ¹⁴CH₄ measurements are more challenging due to a far lower atmospheric concentration of CH₄ compared to CO₂ but also due to potential contamination of CO₂ during CH₄ extraction of atmospheric samples. In this context, a new CH₄ preconcentration and purification setup, which allows ¹⁴CH₄ measurements of atmospheric air was recently developed at the Laboratory for the Analysis of Radiocarbon with AMS (LARA) at the University of Bern (Epic et al., 2019).

Since 2019, three strategic sites in Switzerland have regularly been visited and air samples are collected: the high altitude research station Jungfraujoch (continental background), the Beromünster (LU) tall tower (rural area) and the Department of Chemistry, Biochemistry and Pharmaceutical Sciences in Bern (urban area). Since September 2021, we have also collected samples at the Sottens (VD) tall tower.

In 2022, we continued with the collection of grab samples (duration about 20 mins) of ambient air at the Sphinx observatory at Jungfraujoch every second week within the framework of the Sinergia project “Radiocarbon Inventories of Switzerland (RICH): An integrated approach to understand the changing carbon cycle” that is funded by the Swiss National Science Foundation (SNSF). Being on-site 25 times, we transported back to Bern in total 50 air samples of ~100 L (at STP) transferred in PE-AL-PE bags (TESSERAUX, Germany). Two bags per sampling day were filled to have a backup if one bag was leaking but also to investigate the repeatability of the measurements. In the laboratory, CH₄ and CO₂ were extracted from the air samples using the dedicated setup that

involves a gas-chromatography (GC) separation of the carbon-containing gases (Epic et al., 2019). The ¹⁴C 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).

The ¹⁴CH₄ measurements in 2022 confirmed the results of 2019-2021 that the variability of the individual measurements from Jungfraujoch was much smaller than for Beromünster, Bern and Sottens. Moreover, F¹⁴CH₄ values measured at Jungfraujoch are generally lower than at the three sites in the Swiss Plateau. This result appears contra intuitive at first sight: these three sites are actually closer to CH₄ emissions and these CH₄ sources, whether biological or fossil, should decrease the F¹⁴CH₄ values compared to the F¹⁴CH₄ values measured at a background site like Jungfraujoch. However, we attributed these elevated F¹⁴CH₄ values in the Swiss Plateau to low-level but omnipresent ¹⁴CH₄ releases of nuclear power plants, and especially pressurized water reactors, at the scale of the whole European continent (e.g. Switzerland, France, Germany, Belgium).

Already announced for 2022, but finally postponed due to delivery issues and longer than expected quality assurance tests, we plan for 2023 to set up an automated sampling system at Jungfraujoch that allows a continuous collection of ambient air samples during night periods for an integrated determination of ¹⁴CH₄. This data will serve as reference for regional source apportionment of methane using ¹⁴C that shall be continued at different sites in Switzerland.

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Scientific publications and public outreach 2022

Conference Papers

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Address

Laboratory for the Analysis of Radiocarbon with AMS
Department of Chemistry, Biochemistry and Pharmaceutical Sciences
University of Bern
Freiestrasse 3
CH-3012 Bern
Switzerland

Contacts

Prof. Dr. Sönke Szidat
Tel.: +41 31 684 43 08
e-mail: soenke.szidat@unibe.ch

Dr. Thomas Laemmel
Tel.: +41 31 684 42 65
e-mail: thomas.laemmel@unibe.ch

Internet data bases

<https://www.14c.unibe.ch/>
<http://p3.snf.ch/project-193770>

Collaborating partners / networks

Prof. T.I. Eglinton, Department of Earth Sciences, ETH Zurich
Dr. F. Hagedorn, Swiss Federal Institute for Forest, Snow and Landscape Research WSL, Birmensdorf
Dr. H. Graven, Department of Physics, Imperial College London
Prof. M. Leuenberger, Climate and Environmental Physics, University of Bern
Prof. D. Brunner, Dr. S. Henne, Empa, Dübendorf
Dr. L. Wacker, Laboratory of Ion Beam Physics, ETH Zurich
Dr. S. Hammer, Institute of Environmental Physics, Heidelberg University