Development and First Deployment of an Innovative Airborne $\delta^{13}C(CH_4)$ In Situ Measurement Setup

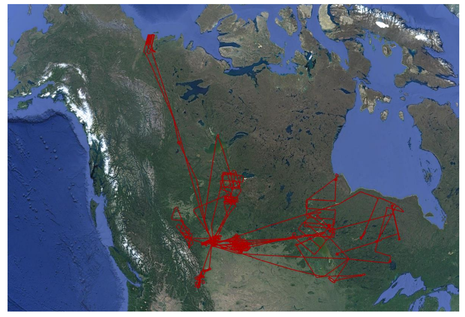
Paul Waldmann (1), Michael Lichtenstern (1), Friedemann Reum (1), Helmut Ziereis (1), Alina Fiehn (1), Michał Gałkowski (2), Christoph Gerbig (2), Andreas Fix (1) and Anke Roiger (1)

(1) Institute of Atmospheric Physics, German Aerospace Center, Oberpfaffenhofen, Germany
(2) (2) Max Planck Institute for Biogeochemistry, Department Biogeochemical Systems, Jena, Germany

Introduction

Recent atmospheric methane concentrations show an accelerated increase, but the contributions of the underlying emitters are poorly understood. Recording the stable carbon isotope ratio of methane ($\delta^{13}C(CH_4)$) is a powerful tool for CH_4 source attribution and the understanding of the global methane budget.
As part of the CoMet 2.0 arctic campaign, which took place in Canada in summer 2022, an innovative, airborne system for the measurement of $\delta^{13}C(CH_4)$ was developed and deployed.

HALO, approaching the runway in Edmonton



The research flights were performed with the High Altitude and Long Range research aircraft HALO and the campaign was based in Edmonton, Alberta.
Various CH_4 emitters are located in Canada:

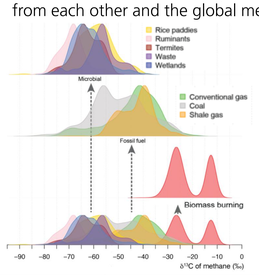
- large scale boreal and arctic wetland areas
- Permafrost regions
- Fossil fuel mining and processing facilities

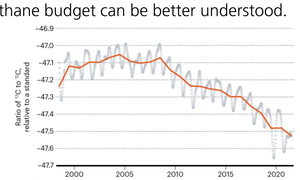
Right: Map of Canada with the flight tracks of HALO during CoMet 2.0

The isotopic ratio of methane sources

- *Biogenic* methane (microbial production):
  - contains little of the carbon isotope ^{13}C.
  - wetlands, waste, rice paddies, metabolism of organic matter by ruminants and termites.
- *Pyrogenic* methane (incomplete combustion):
  - rich in ^{13}C
  - product of incomplete combustion of organic matter.
  - bimodal distribution in the isotopic signature (C3 & C4 plants).
- *Thermogenic* methane:
  - isotope ratios located between pyrogenic and biogenic methane
  - thermal breakdown of organic matter under high temperatures and pressures, i.e. in geological gas deposits.

Based on the different isotope ratios, methane sources can be distinguished from each other and the global methane budget can be better understood.





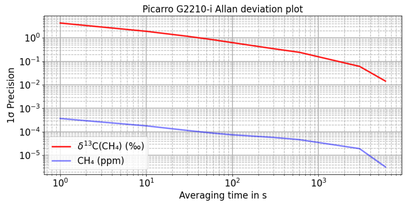
Top: Global atmospheric $\delta^{13}C(CH_4)$ trend from *Jeff Tollefson, nature, 2022*

Left: Density distribution of isotopic signatures from *Sherwood et al., earth system science data, 2017*

The $\delta^{13}C(CH_4)$ measurement system MIRACLE

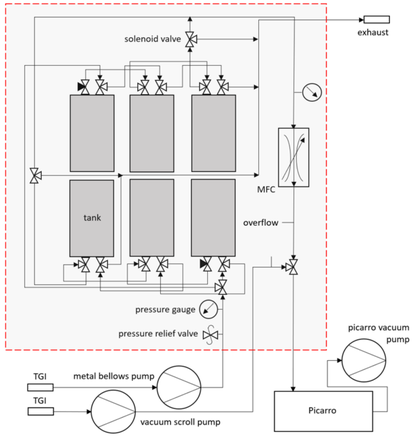
MIRACLE instrument

- Picarro G2210-i greenhouse gas analyzer (cavity ring down spectroscopy).
- dry mole fractions of CH_4, C_2H_6, CO_2 and $\delta^{13}C(CH_4)$.
- vacuum scroll pump: supplies analyzer with ambient air.
- *Novel sampler unit.*



Sampler unit

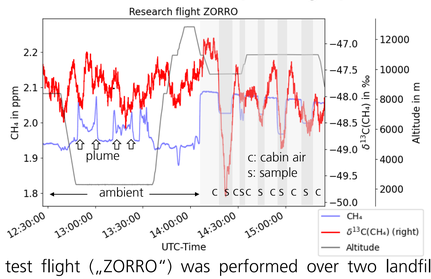
- Additional metal bellows pump.
- Six 2l stainless steel tanks (flush & fill time: 40s, tank pressure: 2.5 bar abs.).
- In flight analysis time up to 10 min per tank → high precision GHG and $\delta^{13}C(CH_4)$ measurements (1σ: 0.34‰ for 5 min)



Flowsketch of the MIRACLE instrument

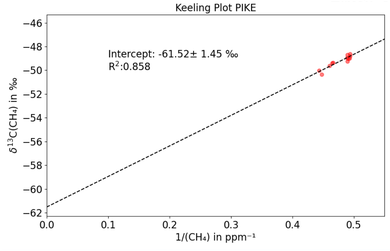
→ *Enables precise meauserement of small scale CH_4 enhancements*

Madrid landfill (test flight)

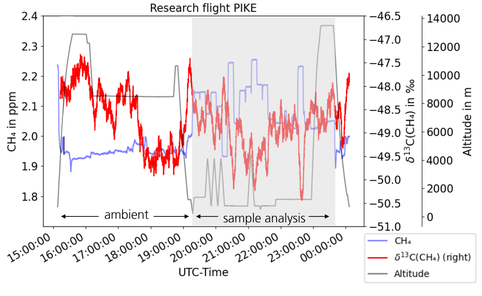


The test flight („ZORRO") was performed over two landfills near Madrid. The plume(⇧) transects were short, with no detectable change in $\delta^{13}C(CH_4)$. The samples (s), taken in the plume and analyzed for several minutes allow for precise $\delta^{13}C(CH_4)$ determination (-83.64 ± 9.51 ‰).

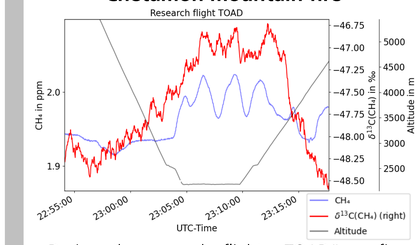
Lake Winnipeg wetlands



The target area of research flight N°9, called „PIKE", were the lake Winnipeg wetlands in Manitoba, Canada. In the first part of the flight, a clear shift in $\delta^{13}C(CH_4)$ indicates different air masses. Low windspeeds the night before and a shallow boundary layer height at the time of the flight caused methane enhancements of 250 ppb compared to the background. Samples containing high CH_4 concentrations showing a shift of $\delta^{13}C(CH_4)$ towards less ^{13}C. The Keeling plot results in a biogenic $\delta^{13}C(CH_4)$ signature of -61.52 ± 1.45 ‰. The result is valid for a large, inaccessible boreal wetland with an extent of several 10000 km².



Chetamon mountain fire



During the research flight „TOAD", a fire plume was spontaneously flown through. HALO crossed into the plume (between 2000 and 3500m) three times. CH_4 and $\delta^{13}C(CH_4)$ are correlated strongly during the profile. The resulting isotopic signature is -22.27 ± 3.7 ‰.

Summary

- innovative instrument, which performs $\delta^{13}C(CH_4)$ measurements in two different modes:
  - continuous from ambient air.
  - semicontinuous when analyzing samples.
- combines high precision, the advantages of airborne deployment and in flight evaluation.
- successfull deployment in the course of the CoMet 2.0 arctic campaign aboard HALO
- $\delta^{13}C(CH_4)$ signatures determined for different anthropogenic and natural methane sources in Canada (wetlands, forest fires, landfill)
- renewed deployment

Deutsches Zentrum für Luft- und Raumfahrt e.V.
Paul.Waldmann@dlr.de

Institut für Physik der Atmosphäre
http://www.dlr.de/ipa