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1. Executive Summary

A lightweight high-precision CH₄ spectrometer based on a guantum cascade laser (QCL) has been developed and deployed on an unmanned aerial vehicle (UAV) at Empa, which will be used to study CH₄ sources in collaboration with Swiss partners and other MEMO² ESRs from Groningen and Lund.

A lightweight active AirCore system that is capable of simultaneous measurements of $CH_4 / CO_2 / CO$ has been developed at the University of Groningen (RUG), which subsequently has been flown aboard a UAV on three individual days downwind of a dairy farm in Grijpskerk in the Netherlands to quantify CH₄ emissions. Besides the profile measurements, the collected air samples were also used for CH₄ isotopic composition measurements by ESRs in Utrecht and London. Furthermore, collaborations with ESRs at Empa and in Wageningen on both field campaigns and modeling work are foreseen.

At this stage, both the lightweight CH₄ spectrometer and the active AirCore system are readily available to perform field measurements.

2. Introduction

2.1 Background

CH₄ emissions have been widely studied by atmospheric measurements on the ground and on piloted aircraft, using primarily laser-based spectrometers, e.g. cavity ring-down spectrometers, or cavity enhanced spectrometers.

Aircraft is one of the most important platforms to perform large-scale vertical and horizontal measurements. However, research aircrafts are not suitable for measurements below ~150 m or during nighttime due to strict aviation regulations for the sake of safety. Small unmanned aerial vehicles (UAVs) has a great advantage to fill the vertical gap of measurements between the surface and the lowest altitude usually reachable by aircraft. Besides this, small UAVs are also cost effective and may be easily deployed.

In recent years, efforts have been made to develop applications using UAVs as a complementary platform for GHG measurements (Table 1).

Platform	Measured species	Application	Reference
Helikite (inlet)	CO ₂ , CH ₄	Monitoring of fossil fuel emissions from a power plant	Turnbull et al., 2014
NASA UAV (40 kg payload)	CO ₂ , CH _{4,} H ₂ O	Characterization of Arctic Sea Ice Experiment	Berman et al., 2012
Robotic helicopter (1-2 kg)	CO ₂ , CH _{4,} H ₂ O	High spatial mapping of local greenhouse gases	Khan et al., 2012
Octocopter	δ ¹³ C in CH ₄	Identification of CH ₄ sources	Brownlow et al., 2016
Fixed-wing & Rotary (inlet)	CO ₂ , CH ₄	Quantification of landfill CH ₄ emissions	Allen et al., 2018

Table 1: overview recent studies using UAVs as a platform for GHG measurements



The available CH_4 spectrometers are either not precise enough or are too heavy to be deployed on a light UAV. To this end, we aim to develop lightweight systems (1-2 kg) for atmospheric measurements of CH_4 mole fractions on UAVs: a lightweight spectrometer and an active AirCore system.

The advantage of the lightweight spectrometer is that it provides in situ high-precision CH_4 measurements that could be easily deployed to identify the location of the emission sources and to further quantify the emission sources. On the other hand, the accuracy of the spectrometer measurements may suffer from environmental variations of temperature, pressure and relative humidity.

Since the active AirCore system collects atmospheric air samples, it has the advantage of making simultaneously high-accuracy measurements of multiple species as well as isotopic compositions of CH_4 . However, the measurements will not be available until the air samples have been analyzed on the ground after landing, which makes it challenging to capture the plumes of emission sources. Furthermore, uncertainties associated with the spatial coordinates of the measurements exist due to possible inaccuracy in the retrieval of the AirCore measurements. Therefore, the two systems could be used as complementary tools in the field to quantify CH_4 emissions.

2.2 Scope of the deliverable

The deliverable includes

- 1) The development of a high-precision CH₄ spectrometer on a UAV platform by Empa
- 2) The development of an active AirCore system on a UAV platform by RUG.

3. Content

3.1 Lightweight CH₄ spectrometer

The lightweight CH_4 spectrometer is based on the absorption of light by the methane molecule. More specifically, a mid-IR source (quantum cascade laser, QCL) emits light which is selectively absorbed by methane in a circular multipass cell. The light is then focused on a very sensitive detector, and the signal treated by fast electronics. The spectrometer thus consists of three main parts: a laser module, a multi-pass cell and a detector unit.

The laser module is a single-mode QCL (Alpes Lasers), housed in a TO-3 package with a Peltier element and collimating optics. Attached to the housing is an external cooling element for enhanced temperature control as well as an ND filter (Thorlabs) to prevent saturation of the detector. A variability in wavelength is introduced by altering the current fed to the laser very precisely, resulting in an emission profile between 1276.5 and 1277.2 cm⁻¹.



D1.1: Lightweight CH₄ sensor and AirCore developed and deployed on UAV

The laser module is directly coupled to the multi-pass cell. The cell is a circular construction with an array of mirrors machined into the inner surface of the cell. The mirrors are angled in such a way that light coupled into the cell makes multiple passes through the volume contained by the cell before being coupled to the cell output, as shown in Fig. 1. By increasing the path length, the absorbed signal increases and improves the signal-to-noise ratio (SNR). The current path length through the cell is 9.92 m.



Fig. 1: Left: A sketch of the working principle of the multi-pass cell. The laser beam enters the cell and is reflected multiple times through the cell volume to enhance SNR. Right: The cell in action. As the beam travels further through the cell, the intensity visibly decreases due to continuous absorption and scattering in the cell volume.

The detector unit consists of a single-pixel detector (Vigo System S.A) and a focusing lens (Thorlabs) that focusses the beam onto the detector pixel. A short distance is left between the lens and the multi-pass cell in order to fit an etalon for frequency calibration. The aperture is mounted on a base plate together with electronics for controlling the laser and handling data acquisition, sensors for pressure, temperature and relative humidity for real-time adjustment of environmental parameters, as well as communication ports for wireless communication devices and system battery. This is displayed in Fig. 2. The total weight of the system, including a battery, is 2.1 kg.



Fig. 2: The spectrometer array mounted on the base plate with electronics and environmental sensors.



D1.1: Lightweight CH₄ sensor and AirCore developed and deployed on UAV

The system is open-path with the CH_4 mole fraction being continuously monitored inside the cell volume. Within the emission band of the laser system are two absorption peaks, one for CH_4 and one for H_2O (also used for frequency locking), as displayed in Fig. 3.



Fig. 3: The transmission profile of ambient air in the wavelength band of interest is displayed in black, together with its constituent absorbing molecular transitions. Two absorption peaks are present, one for H_4 and one for H_2O . The N_2O peak shown in this spectrum is present but outside the scanning bandwidth of the laser.

The laser is scanned over the tuning bandwidth once every millisecond. In order to limit the required bandwidth of the wireless connection between the device and control station, as well as the computational power needed for spectral fitting, N spectra (N typically 1000) are averaged before being saved and sent from the device to the control station.

Parts-per-billion (ppb) precision has been achieved in laboratory environment, with the best observed precision being significantly 0.1 ppb, as shown in Fig. 4.



Fig. 4: Top: The detected CH_4 mole fraction in laboratory environment. Bottom: The Allan Variance plot showing the excellent precision of the system. The black line shows typical precision and the green line shows the best observed precision. The diagonal grey dashes indicate the 1/t limit while the horizontal grey dashes indicate 1 ppb precision. Spectra were measured at 1 kHz, i.e. 1 s integration time corresponds to the averaging of 1000 spectra.



D1.1: Lightweight CH₄ sensor and AirCore developed and deployed on UAV

The system has been used in the field, mounted on a DJI Matrice 600 UAV. The UAV was flown 4-7 m over a field where a stable CH_4 source had been placed at a height of 5 m. The detected increased CH_4 mole fraction over ambient is displayed in Fig. 5Fig, overlaid with a map of the area and the mole fraction increase predicted by simulations using a Lagrangian particle model (GRAL).



Fig. 5: The map of a field outside the Empa facility in Dübendorf. A stable CH_4 source was placed 5 m above ground. The increase in CH_4 mole fraction above average was simulated using a GRAL model. The dots indicate the increase measured by the UAV instrument.

3.2 The active AirCore system

3.2.1 development of the active AirCore system

The active AirCore consists of ~50 m stainless steel tubing with an O.D. of 3.175 mm (1/8 in.) and wall thickness of 0.127 mm (0.005 in.) a dryer, a micropump, and a datalogger. The inner surface of the tubing is coated with SilcoNert 1000 by Restec Inc. To obtain a constant flow through the AirCore, an orifice (O.D. $\frac{1}{4}$ in., orifice diameter 45 ± 10% µm, Lenox laser Inc.) is placed between the pump and the coiled tube. The system is placed in a carbon fiber box and attached to the UAV using two carbon fiber rods. The total weight of the active AirCore system box is ~1.1 kg (see Fig. 6, Andersen et al., 2018).





Fig. 6: (a) schematic design of the UAV AirCore system. (b) image of the UAV AirCore system

Prior to every flight, the active AirCore is flushed with a calibrated fill gas that is spiked with ~ 10 ppm CO, which helps to identify the starting point of ambient air sampling during later analysis. The active AirCore starts to collect air samples when the micropump is turned on using a switch located outside the box shortly before a UAV flight, and the pump is turned off manually after the UAV lands. Air samples are collected during the flight and retained within the active AirCore. The active AirCore samples are then immediately analyzed with a trace gas analyzer (CRDS, Picarro, Inc., CA, model G2401).

The active AirCore system was flown nearby the 60 m tall Lutjewad tower to compare measurements of the drone flights with data obtained from the tower monitoring. Fig. 7 shows the vertical profiles of CH_4 , CO_2 , and CO from three consecutive UAV flights in the same morning. The vertical profiles captured the mixing of the surface layer with air aloft. The active AirCore measurements at 60 m and 7 m agree well with the measured mole fractions at 60 m and 7 m at the tower.





Fig. 7: vertical profiles of a) CO_2 , b) CH_4 and c) CO for flight nos. 1-3. Figures a) and b) include a dotted line indicating 60 m and shows measured trace gas mole fractions from the Lutjewad atmospheric station at this height. Figure b) include also a dotted line to indicate 7 m height and the corresponding CH_4 values obtained from the atmospheric station at this height. The square points represent the mole fractions measured at the time of the UAV ascent, and the circular points represent the mole fractions measured during the UAV descent. The color of the markers represents its respective flight. The CO mole fractions shown in (c) has been averaged by every fifth data point, with each dot representing a data point with a time resolution of 3 seconds.

3.2.2 Application of the active AirCore system to quantify CH₄ emissions from a dairy farm in Grijpskerk, the Netherlands

UAV AirCore measurements of CH₄ have been made on three days in March 2017, May and October 2018 downwind of a dairy cow farm, ~ 20 km North–West of the city of Groningen, which was chosen because it is one of the largest dairy farms in the Netherlands. The CH₄ mole fraction measurements from the UAV AirCore (8 – 15 minutes) were used to determine the CH₄ enhancement of the downwind against the upwind.





Fig. 8: CH₄ mole fraction, March 27th 2017. The wind came from the South – East. (a) Upwind. (b) Downwind

To integrate the enhanced CH_4 mole fraction, data needs to be interpolated since it is unequally distributed in space. A three-dimensional plot showing the spatial distribution of measured CH_4 mole fractions, together with interpolated data (Fig. 9c), which were used to estimate the flux with a mass balance method.



Fig. 9: 3D plot of CH₄ mole fraction, March 27th 2017: (a) Upwind. (b) Downwind. (c) Interpolated data.



4. Conclusion and possible impact

The lightweight CH_4 spectrometer and the lightweight active AirCore system have been successfully developed and deployed on UAVs, which enables us to perform state-of-the-art high-precision atmospheric measurements of CH_4 to quantify various hot-spot emissions, e.g. from dairy farms, rivers, landfills, wetlands.

The two available measurement systems will be deployed within the MEMO² project to generate atmospheric measurements downwind of various CH_4 emission sources. The dataset will be useful to validate model simulations in the same consortium. The combination of observations and modeling work would allow us to better quantify the emission strength.

5. Dissemination & Exploitation

1. Huilin Chen, Truls Andersen, Katarina Vinkovic, Marcel de Vries, Bert Kers, Wouter Peters, Chiel van heer Waarden, Anja Raznjevic, Maarten Krol, Thomas Röckmann, Malika Menoud, Jarosław M. Nęcki and Anke Roiger: Quantifying Hotspot Methane Emissions Using a UAV-based Active AirCore System, American Geophysical Union, Fall Meeting 2018

2. **Katarina Vinkovic**, Truls Andersen, Marcel de Vries, Wouter Peters, **Arjan Hensen**, **Huilin Chen**: Quantification of methane emissions from dairy cows in the Netherlands, The 3rd ICOS Science Conference, Prague, 2018

Bolded names are MEMO2 members.

6. References

Allen, G., Hollingsworth, P., Kabbabe, K., Pitt, J. R., Mead, M. I., Illingsworth, S., Roberts, G., Bourn, M., Shallcross, D. E., and Percival, C. J.: The development and trial of an unmanned aerial system for the measurement of methane flux from landfill and greenhouse gas emission hotspots, Waste Management, https://doi.org/10.1016/j.wasman.2017.12.024, 2018.

Andersen, T., Scheeren, B., Peters, W., and Chen, H.: A UAV-based active AirCore system for measurements of greenhouse gases, Atmos. Meas. Tech., 11, 2683-2699, https://doi.org/10.5194/amt-11-2683-2018, 2018

Berman, E. S. F., Fladeland, M., Liem, J., Kolyer, R., and Gupta, M.: Greenhouse gas analyzer for measurements of carbon dioxide, methane, and water vapor aboard an unmanned aerial vehicle, Sensors and Actuators B: Chemical, 169, 128-135, doi: 10.1016/j.snb.2012.04.036, 2012.

Brownlow, R., Lowry, D., Thomas, R. M., Fisher, R. E., France, J. L., Cain, M., Richardson, T. S., Greatwood, C., Freer, J., Pyle, J. A., MacKenzie, A. R., and Nisbet, E. G.: Methane mole fraction and $\delta 13C$ above and below the trade wind inversion at



Ascension Island in air sampled by aerial robotics, Geophysical Research Letters, 43, doi: 10.1002/2016GL071155, 2016.

Khan, A., Schaefer, D., Tao, L., Miller, D. J., Sun, K., Zondlo, M. A., Harrison, W. A., Roscoe, B., and Lary, D. J.: Low Power Greenhouse Gas Sensors for Unmanned Aerial Vehicles, Remote Sens., 4, 1355–1368, https://doi.org/10.3390/rs4051355, 2012.

Turnbull, J. C., Keller, E. D., Baisden, T., Brailsford, G., Bromley, T., Norris, M., and Zonder van, A.: Atmospheric measurement of point source fossil CO2 emissions, Atmospheric Chemistry and Physics, 14, 5001-5014, doi: 10.5194/acp-14-5001-2014, 2014.

7. History of the document

7.1 Document history

Version	Author(s)	Date	Changes
Version 0.1	Huilin Chen	23.01.2019	Overall and RUG input
Version 0.2	Lukas Emmeneg- ger/Jonas Ravelid	13.02.2019	Empa input
Version 1.0	Huilin Chen	13.02.2019	A complete draft

7.2 Internal review history

Internal Reviewer	Date	Comments
Sylvia	25.02.2019	See above
Sylvia	06.03.2019	Deliverable report as brief summary of task 1.3, not a detailed description of de- velopment work as the re- port is public and technical details will be published as a scientific paper