Airborne Lidar Measurements of Anthropogenic Methane Sources with CHARM-F during the CoMet 2.0 Arctic Campaign

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Abstract: The airborne Integrated Path Differential Absorption Lidar CHARM-F for accurate column concentration measurements of CH_4 and CO_2 has been very successfully deployed on a HALO aircraft campaign over Canada in late summer 2022. The scientific target areas, apart from natural sources (wetlands and thawing permafrost areas), included a variety of anthropogenic sources (oil and gas industry, coal mining, landfills and power plants). Using selected examples, we show the capabilities, but also the challenges, of trying to quantify methane fluxes from such measurements.

1. The CHARM-F IPDA Lidar

CHARM-F (CH4 and CO2 Atmospheric Remote Monitoring – Flugzeug) is an airborne lidar system for simultaneously measuring column concentrations of CO_2 and CH_4 between ground and flight level [1,2]. The system is designed to be operated onboard the HALO (High Altitude and Long Range) aircraft with the transceiver unit facing downward through optical windows at the bottom of the aircraft fuselage. The transmitters are based on optical parametric oscillators (OPOs) which are pumped by 50-Hz double-pulse Nd:YAG lasers and injection seeded by cw DFB lasers. The OPOs operate at 1572 nm and 1645 nm respectively, with energies of about 10 mJ/pulse.

CHARM-F employs the integrated path differential absorption (IPDA) technique [1,2], which uses pulsed lasers at two precisely defined wavelengths – one partially absorbed, the other largely unaffected by the targeted trace gas – for measuring the average concentration within the column below the aircraft from the intensity backscattered at the ground. The method is illustrated in Figure 1, which also shows the concept of emitting double pulses at close intervals to maximize the overlap of illuminated points on the ground. The quantity that is directly measured with IPDA is the so-called differential-absorption optical depth

$$DAOD = \frac{1}{2}\ln\left(\frac{S_{\mathrm{off}}/E_{\mathrm{off}}}{S_{\mathrm{on}}/E_{\mathrm{on}}}\right), \tag{1}$$

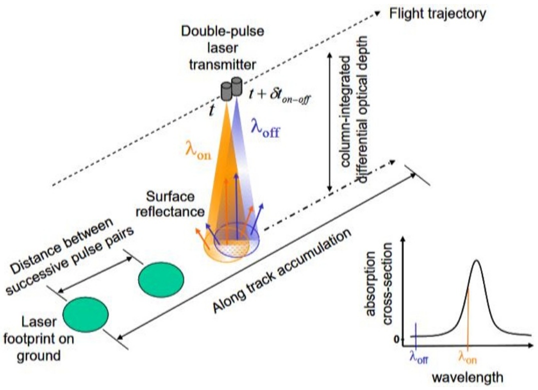

Figure 1. Illustration of the IPDA measurement technique. Illustration from [2]

where S_{on} and S_{off} are measurements of the backscattered signal and E_{on} and E_{off} are the internal energy reference measurements. The weighted column averaged dry-air mixing ratio

$$X_{\mathrm{GHG}} = \frac{DAOD}{\int_{p_{\mathrm{plane}}}^{p_{\mathrm{ground}}} WF(p,T)\cdot \mathrm{d}p} \tag{2}$$

is computed using a pressure- and temperature-dependent weighting function that considers the precise spectral properties of the trace gas,

$$WF(p,T) = \frac{\sigma_{\mathrm{on}}(p,T) - \sigma_{\mathrm{off}}(p,T)}{g\cdot\left(m + m_{\mathrm{H_2O}}\cdot r_{\mathrm{H_2O}}\right)}. \tag{3}$$

The absorption cross-sections $\sigma_{\mathrm{on}}(p,T)$ and $\sigma_{\mathrm{off}}(p,T)$ are taken from the HITRAN database [5]. g is the gravitational acceleration, m and $m_{\mathrm{H_2O}}$ are the average molecular mass of dry air and water vapor respectively and $r_{\mathrm{H_2O}}$ is the

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mixing ratio of the latter, taken from weather models.

The cross-sectional greenhouse gas mass flux q can directly be computed from the enhancement in $DAOD$, that is associated with the forming plume. Assuming that the approximate altitude of the plume is known, the enhancement with respect to the background $\Delta DAOD$ is integrated along the plume-crossing flight track s [3,4].

$$q = \int \Delta DAOD\, ds \cdot \frac{M_{ghg}}{\Delta\sigma} \cdot u^{\perp}. \tag{4}$$

M_{ghg}, $\Delta\sigma$ and $u^{\perp}$ are the molar mass, the differential molar absorption cross-section and the effective wind speed across-track, respectively.

2. The CoMet 2.0 Arctic campaign

The CoMet 2.0 Arctic airborne campaign, scheduled between August and September 2022, and based in Edmonton Alberta, Canada. It had the goal of measuring the concentrations and identifying the sources of greenhouse gases in the Canadian boreal wetlands and at sites of human activities. The measurement flights for CoMet 2.0 Arctic were carried out onboard the German research aircraft HALO. The CHARM-F lidar was operated as part of a comprehensive suite of instruments, comprising active and passive remote sensing, as well as in-situ instruments. Figure 2 shows an overview of all ground tracks during the research flights that were flown out of Edmonton, for which good-quality CHARM-F measurements are available. In addition, measurements were conducted during the two transfer flights, as well as during the scientific test flight to landfills in the Madrid area in Spain, flown out of Oberpfaffenhofen, Germany. Apart from the natural source areas of methane, typically located further away from the campaign base, in the vast wetland and permafrost areas south and west of the Hudson Bay, as well as in the Mackenzie river delta, at the Arctic coast, most of the shorter flights were targeting locations of antropogenic activities, mostly associated with fossil fuel extraction.

3. Towards flux measurements from anthropogenic sources

The portfolio of anthropogenic sources of carbon dioxide and methane probed during the CoMet 2.0 Arctic campaign comprises a coal-fired power plant near Edmonton, oil and gas extraction sites, open-pit coal mines, as well as landfill sites, both in Canada and Spain. The good precision and high accuracy of CHARM-F column measurements enabled detecting and localizing as well as quantifying the emissions from a variety of different targets.

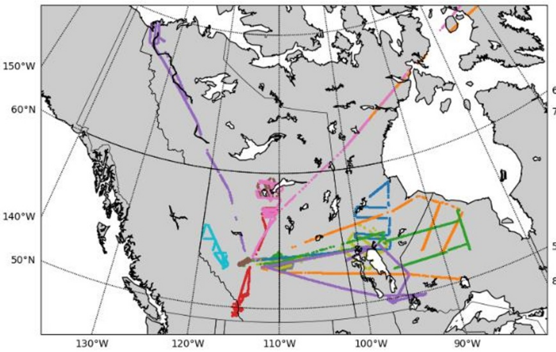

Figure 2. All CHARM-F ground tracks of CoMet 2.0 Arctic over Canada (data-quality filtered).

3.1. Methane plumes from Madrid landfills

The first interesting case that is explored here in some detail are methane plumes from the landfills of Valdemingomez and Pinto near Madrid, Spain, that were targeted during the scientific test flight of the campaign on August 4th, 2022. This particular measurement is interesting, as it serves as independent validation of satellite- and ground-based measurements that already identified the landfills as strong methane sources, giving flux estimates of 7100 kg/h for both sites (TROPOMI) and 3500 kg/h for the Valdemingomez site alone (COCCON) [6]. These CHARM-F measurements not only revealed a well-developed plume downwind, north of both landfills, enabling cross-sectional flux measurements, but also a region of residual nightly accumulation of methane to the west (see Figure 3). Estimates of the emission rate, based on the cross-sectional flux method (Equation 4) result in a combined emission rate of 9800 ± 3100 kg/h for both landfills. This measurement is obtained by selecting all flight tracks that cross the merged plume down-wind of both landfills (to the north-east). An average wind speed of 4.3 m/s was assumed, which was obtained by averaging ERA5 reanalysis data for the location of the plume, the time of the flight

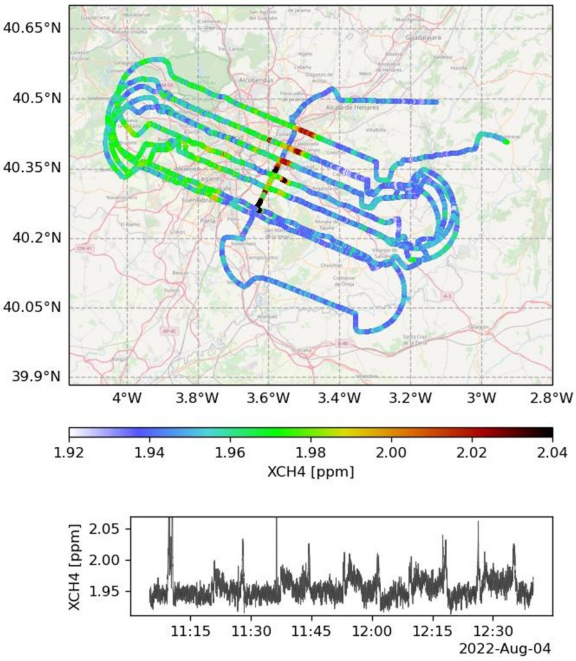

Figure 3. Methane column mixing ratio along flight-tracks over Madrid, Spain. Top: color coded concentration along ground track. Bottom: Mixing ratio over flight time (UTC).

and across the, at local noon, highly turbulent boundary layer. For each of the plume transects $\Delta DAOD$ is obtained by using a window, centered on the in-plume enhancement and adjacent windows of the same length before and after the plume for estimating the background. These windows are varied between 4 km and 8 km in width to estimate the uncertainty of this approach.

The result that is obtained in this example, due to the well-developed plume, can be regarded as a reasonably good estimate of the actual emission rate, especially in light of the rather conservative error estimate, which also considers the spatial and temporal variability in the wind field. The wind history indicates a rotation of the wind direction in the early morning, linking the enhanced concentration over the city (left side in Figure 3) to a residual plume or nightly accumulation. More accurate estimates of the methane flux in such a scenario can be achieved by supporting the observations with high-resolution weather and tracer simulations at local level [7].

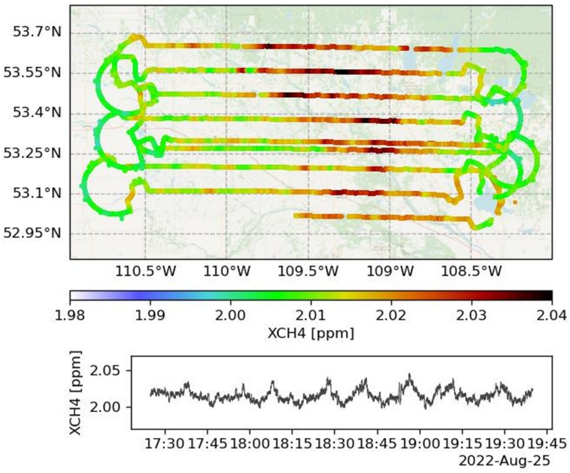

Figure 4. Methane column mixing ratio along flight-tracks over the Lloydminster oil and gas fields, Canada. Top: color coded concentration along ground track. Bottom: Mixing ratio over flight time (UTC).

3.2. Diffuse methane plume over the Lloydminster oil and gas filed

Another example of a measurement, for which the high accuracy of CHARM-F plays a crucial role has been conducted on August 25th, 2022 over the oil and gas extraction sites close to Lloydminster at the border between Alberta and Saskatchewan, Canada. The region exhibits numerous individual oil/gas wells, each with some potential for methane leakage, but mostly too weak for detection on a single source level [8]. However, a plume-like concentration enhancement on the level of about one percent has been detected over the region (see Figure 4). Applying the cross-sectional flux method here yields 24 ± 8 t/h as an average over all plume transects. For this rough estimate, the boundary-layer wind speed is averaged over 10 hours prior to the end of the measurements. The width of the plume is varied between 40 and 60 km.

It is obvious that for this scenario the cross-sectional flux method reaches its limit. This is mainly due to the long timescales needed for the air parcels to traverse the probed region and the variability in the wind field over such a long timescale. Another aspect that cannot easily be probed with this method is the spatial distribution of sources that have to be assumed as scattered over a rather large area. Major improvements are to be expected when attempting a model inversion approach that is based on regional weather simulations [7].

3.3. Methane accumulation over the open-pit coal mines in the Rocky Mountains

The last example highlights the strongest column-enhancements ever observed with the CHARM-F instrument. During the research flight on September 10th HALO targeted open-pit coal mines near Elkford and Sparwood, BC. The mines are located along a valley in the Rocky Mountains, at altitudes between 1500 m and 2000 m a.s.l., with the surrounding peaks reaching up to more than 2500 m. Coal mines are known to be a major contributor to global anthropogenic methane emissions. especially the type of hard coal (Anthracite) that is mined near Elkford is known to release large amounts of methane [9]. Figure 5 shows methane column concentrations along the flight tracks. The region with the highest enhancement is directly located above the Elkford mining site. Column concentrations in some locations reach up to 5000 ppb, more than 2.5 times the background. Despite such a strong signal it is difficult to estimate the source flux, mainly due to the complex topography and unknown wind situation. High resolution weather simulations could help inferring source fluxes also for this data set.

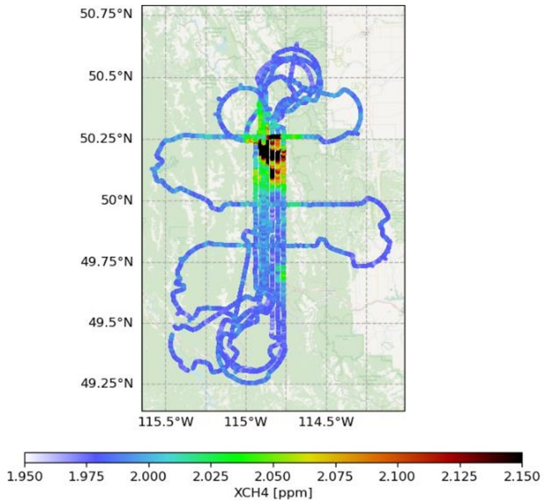

Figure 5. Methane column mixing ratio along flight-tracks over Elkford, BC, Canada.

4. Conclusion

The examples highlighted above demonstrate the excellent performance and high sensitivity of CHARM-F, resolving column enhancements of methane down to the sub-percent level. This allows to probe a variety of potential sources of the important greenhouse gas. The main challenge when attempting flux measurements, especially of highly extended sources or over complex terrain, lies in the uncertainty and variability of the wind field. Local weather simulations and flux inversion approaches show high promise to improve future greenhouse gas flux attribution.

5. Acknowledgements:

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