

Automated CO₂ Profiling with Scanning Raman Lidar in the Lower Troposphere

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Abstract: We have extended our automatic and continuously measuring ground-based Raman lidar ARTHUS (Atmospheric Raman Temperature and Humidity Sounder) with a CO₂ channel. A narrow-band interference filter extracts the 2v₂ CO₂ Raman line with 68 % peak transmission and 0.15 nm FWHM. We use a frequency-tripled Nd:YAG laser (200 Hz, 40 mJ in 2023, 100 mJ in 2024) and a 40-cm receiving telescope. With the current setup, we profile CO₂, H₂O and temperature as well as particle extinction coefficient and particle backscatter coefficient. We have operated this eye-safe system successfully in 2023 for several weeks at our university and continue to do so with a further improved system performance this spring. The first test measurements in 2023 achieved already uncertainties of <1.3 ppm at 1 km altitude with averaging of 4.4 h and 500 m at night.

1. Introduction

CO₂ is one of the most important greenhouse gases but still little is known about its concentration distribution, neither in spatial nor in temporal sense. As the strongest short-term variability occurs near the land surface, ground-based lidar systems could become particularly well suited for range-resolved CO₂ measurements. Since the first lidar measurements of CO₂ by Riebesell et al. in the 1980s [1,2] advances in technology, especially high-transmission narrow-band interference filters and high-power lasers with narrow spectral output, made it possible to isolate the weak signal of carbon dioxide from the close stronger vibrational Raman line of O₂ as well as the rotational-vibrational Raman spectrum of

O₂. Whiteman et al. [3] demonstrated already in 2006 that high-precision measurements of CO₂ with Raman lidar are possible with a frequency-tripled Nd:YAG laser as transmitter. But, to the best of our knowledge, continuous measurements over long periods of time are still missing as well as scanning lidar measurements. To provide this kind of measurements, we developed and integrated a new channel into our automated Raman lidar system ARTHUS (Atmospheric Raman Temperature and HUmidity Sounder). This system has proven to provide high resolution measurements up to the turbulent scale for quantities like temperature and water vapor mixing ratio continuously and fully automatic [4].

2. Setup and Techniques

A schematic setup of the system is given in Fig. 1. We use a wavelength of 355 nm from a frequency tripled Nd:YAG laser (InnoLas) as primary wavelength. The two other wavelengths are separated with the use of a Pellin-Broca prism and absorbed by a beam dump (BD) in order secure eye-safe operation. In the UV, a maximum pulse energy of 100 mJ is available. To achieve eye safety the beam is expanded before a beam steering mirror transmits the beam coaxially with the receiver telescope's optical axis. The back-scattered light is collected by the latter, coupled into an optical fiber and guided to the polychromator. There the beam is first collimated and then

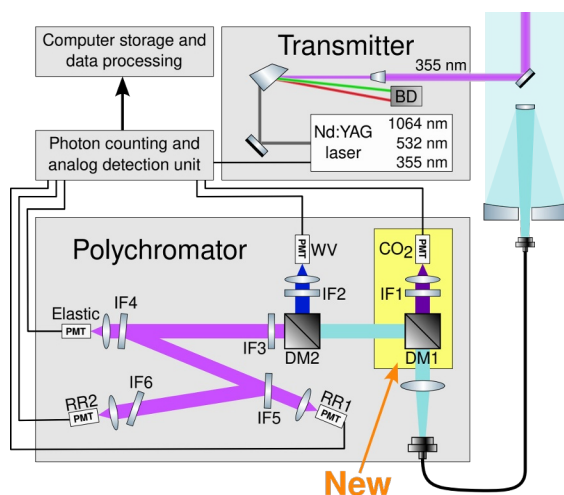


Figure 1: Schematic setup of the instrument.

Table 1: Lidar System Parameters

Symbol	Parameter	Value
E_0	Pulse energy	100 mJ (40 mJ in 2023)
λ	Wavelength	355 nm
f_{rep}	Repetition frequency	200 Hz
d_{tel}	Telescope diameter	400 mm
FOV	Field of view	0.31 mrad
T_{IF}	Peak Transmission of CO ₂ IF	68 %

distributed into five channels by a set of dichroic mirrors (DM) and interference filters (IF). Isolation of the CO₂ 2v₂ Raman line at

372 nm is achieved by a narrow-band interference filter (Materion) with FWHM of 0.14 nm and a peak transmission of 68 %. This narrow FWHM is needed to avoid large contributions of the nearby strong O₂ Raman lines (separation of the 2v₂ CO₂ line to the Q-branch of the vibrational line of O₂ is only 3.8 nm). By angle-tuning of the filter, the central wavelength of the passband was matched to the CO₂ Raman line [5]. The filtered photons are then focused onto photomultiplier tubes (PMT, Hamamatsu) and counted with transient recorders (Licel) in analog and photon counting mode simultaneously. For a discussion of the other channels please see Ref. [4].

Profiles of the CO₂ mixing ratio $M(r)$ are obtained by dividing the CO₂ signal P_{CO_2} by the rotational Raman signal P_{RR1}

$$M(r) = C \frac{P_{CO_2}(r) T_{RR1}(r)}{P_{RR1}(r) T_{CO_2}(r)}$$

where C is a calibration constant obtained, e.g. with an independent ground-based reference measurement. P_{RR1} is used as the reference signal as it contains scattering of all air molecules. The last term is the ratio of the atmospheric transmissions at the corresponding wavelengths of the CO₂ signal $T_{CO_2}(r)$ and the rotational Raman signal $T_{RR1}(r)$. This ratio is not exactly 1 due to the different wavelengths of the signals nor is it constant. There are two contributions, molecular and particle extinction. The former we approximate with the use of the US standard atmosphere. At, e.g., 4 km this reduces the mixing ratio by 4 % compared to the uncorrected signal. The different transmission

due to the particle extinction is more challenging to determine due to the strong variability of particles in altitude, time, size, shape and chemical composition. In principle, the extinction coefficient can be extracted from the rotational Raman signal [2]. We are currently working on the implementation and will show improved profiles at the conference. It should also be noted, that the rotational Raman spectrum shows a non-negligible temperature dependence. A combination of the two rotational Raman signals can be used to get a temperature-independent reference signal [6].

To deal with the high dynamic range of the rotational Raman signal in the near range, we merge the result with the photon count signal at long range and convert this way the analog signal to virtual photon counts. For the weak CO₂ signal the dynamic range in the photon counting mode is sufficient for all ranges.

3. Measurements and Performance

In 2023, we operated the new system successfully for several weeks at the Land-Atmosphere Feedback Observatory (LAFO) [7] of the University of Hohenheim, Stuttgart, Germany. At this time, the pulse energy was about 40 mJ at 200 Hz repetition frequency. For the next measurements 100 mJ will be available. In Fig. 2, we present as example of the first measurement period, a profile of the

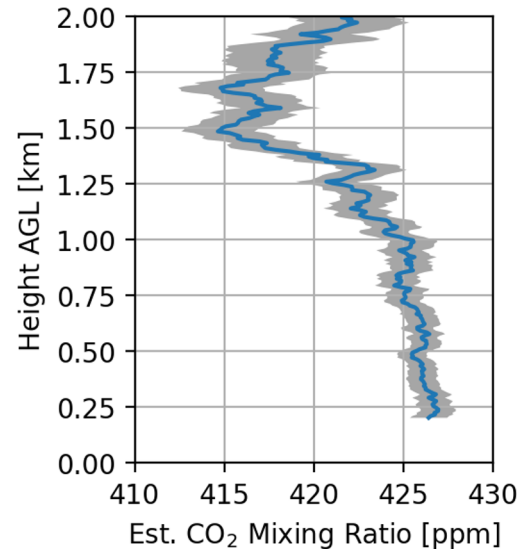


Figure 2: Estimated CO₂ mixing ratio (blue) with precision (gray) from measurements acquired in the night from 2 to 3 November 2023. The averaging is 4.4 h in time and 500 m in range.

CO₂ mixing ratio up to 2 km. For this, signals from the night of 2 to 3 November 2023 where averaged over a cloud-free period of 4.4 h. The calibration constant was determined by assuming a constant mean mixing ratio of 417 ppm above 4 km. This, of course, only provides an approximation of the absolute values while the determination of the uncertainty is still very good. In the future, we will calibrate the measurements with the data of ground-based in-situ instruments at LAFO. For estimating the precision, we assume Poisson statistics. In Fig. 2 the precision is represented as the gray area behind the profile. In Fig. 3 the same precision is presented as a curve over altitude for better readability of the absolute values. The minimum uncertainty of 0.8 ppm appears at 500 m altitude. The uncertainties are < 1 ppm between 250 and 750 m. At 1 km uncertainties of 1.3 ppm are achieved and at 1.5 km still 2 ppm.

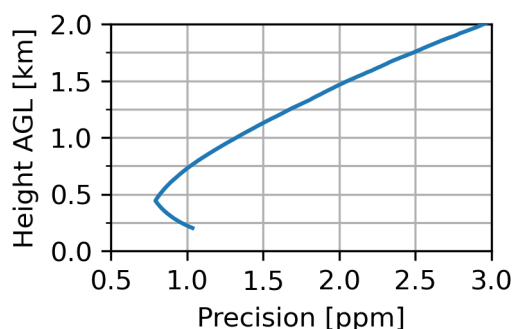


Figure 3: Estimated absolute precision of the measurement presented in fig. 2.

To obtain a realistic estimate of the time and range average needed for 1 ppm in precision at different altitudes and with the future laser pulse energy of 100 mJ we have used the CO₂ and reference signal that were also used to obtain the profile presented in Fig. 2 as reference signals. The resulting curves are presented in Fig. 4. For example, measurements with < 1 ppm uncertainty are expected with a high range resolution of only 60 m at 500 m altitude if the measurement time is 10 h. Alternatively, higher temporal resolution can be traded with larger range averaging, getting the same uncertainty at 500 m for 600 m range and only 1 h time averaging.

4. Conclusion and Outlook

Here we have presented analyses of the first CO₂ measurements with our automated eye-safe

Raman lidar ARTHUS discussing profiles up to 2 km in altitude. With the available laser pulse energy of only 40 mJ in 2023, we already achieved a precision better than 2 ppm up to 1.5 km in altitude when averaging the data over 4.4 h in time and 500 m in range. With the now available 100 mJ pulse energy since this year, only approximately 40 % of the averaging time is needed to achieve the same precision. In the near future, we will expand the system with a scanner unit. This will enable us to precisely calibrate and compare the CO₂ lidar data with in-situ instruments located at the ground. Furthermore, the identification and quantification of carbon sources and sinks along the surface will then be possible.

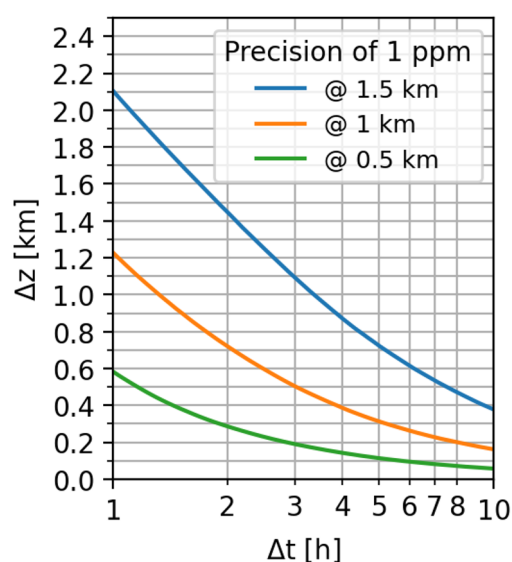


Figure 4: Time and range averaging needed for 1 ppm precision for different altitudes for a pulse energy of 100 mJ.

5. References

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