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A portable laser spectrometer for airborne and ground-based remote sensing of geological CO₂ emissions

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LARSS, a 24 kg, suitcase-sized, CW-laser remote-sensing spectrometer with a ~2 km range has been developed. It demonstrated its flexibility in measuring both atmospheric CO₂ from an airborne platform and terrestrial emission of CO₂ from a remote mud-volcano, Bledug Kuwu, Indonesia from a ground-based sight. This system scans the CO₂ absorption line with 20 discrete wavelengths as opposed to the typical two wavelength online offline instrument. This multi-wavelength approach offers an effective quality-control, bias-control and confidence-estimate of measured CO₂ concentrations via spectral fitting. The simplicity, ruggedness and flexibility in the design allows for easy transportation, use on different platforms with quick setup in some of the most challenging climatic conditions. While more refinement is needed, the results represent a step-stone towards widespread use of active one-sided gas remote sensing in the Earth sciences.

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Geological gases such as SO₂ and CO₂ being linked to subsurface processes are quantitative indicators of Earth's geochemical and geomechanical activity [1-4]. Magmatic CO₂, for instance, is a viable tracer of periods of volcanic unrest [5-7]. As a geological gas, volcanic CO₂ is part of the global geochemical carbon cycle [8,9]. Also part of this cycle is mud volcanism, a manifestation of liquefied sediments and gases reaching the surface due to overpressure and buoyancy [10]. To enhance our understanding of volcanoes and mud volcanoes and their significance within the carbon cycle, measuring their CO₂ output is therefore imperative.

In-situ probing of CO₂ is common [4, 11, 12], but requires knowledge about vent location which may render emission measurements incomplete [7]. Many geological sources emit diffusively, making in-situ probing a lengthy procedure. Particularly near volcanoes and mud volcanoes in-situ measurements may be hazardous to personnel and equipment. In addition, many sites are located in remote and hard to reach areas.

Optical remote sensing offers a spatially comprehensive measurement with large spatial coverage, a safer measurement distance and increased timeliness of acquisition. Passive techniques have demonstrated this for decades [13,14]. Active techniques allow more flexible acquisition geometries. By performing an angular scan, gas concentration profiles are attained, which can be used to compute fluxes. As single-ended, active remote sensors, LIDAR instruments based on differential absorption (DIAL) are particularly viable for that task. They acquire range resolved [15,16] or path averaged [17,18] CO₂ concentrations, the latter using a topographic target (hard target) as backscatterer. Only recently, however, volcanic CO₂ has successfully been measured using these techniques [18,19].

Remote sensing instruments for geological gas detection should be as rugged, compact and portable as possible. Technology has advanced sufficiently for DIAL-type instruments to be realized that fulfill these requirements [20,21]. Yet, these instrumental platforms are still far from mature, but subject to intense research.

Our previous experience shows that inevitable wavelength dependencies of the transmittance of the instrument's optical components itself can make it demanding to differentiate between changes in the baseline due to actual gas absorption and parasitic changes due to non-stationary noise [22,23]. A major contribution to this non-statistical noise is by interference fringes occurring at optical interfaces within the instrument itself. Furthermore, for shorter measurement ranges, where speckle integration over the telescope field of view is less efficient, hard target speckle contribute to non-random noise.

Another major concern for a geological field kit is maximum platform independence: The instrument should be able to perform a scan on the ground and from an airplane in the same campaign.

These issues along with previous experiences inspired the development of a laser remote sensing spectrometer (LARSS), which includes the following key measures:

- increasing the number of wavelengths from 2 to 20,
- adapting efficient data acquisition and processing for multi-platform use,
- increasing the seed laser line width to decrease coherence,
- avoiding transmissive optical elements for reference power measurement.

The resulting instrument is detailed in Fig. 1, Table 1 and is described in the following.

Table 1. Main parameters of LARSS.

Optical power	1 W (CW)
Seed modulation frequency	10 kHz
Sweep rate	20 Hz (variable)
Sweep wavelength range	1572.28 nm to 1572.42 nm
Beam divergence	1.7 mrad
Telescope	200 mm Schmidt-Cassegrain
Focal length	1800 mm
Detector	InGaAs PIN, 2 mm aperture
Digital to analog converter	24 bit @ 50 kS/s
Controller	NI cRio 9076
Power consumption	45 W
Total weight	24 kg

Fig. 1. Overview of LARSS. (a) Basic schematic of LARSS, with wavelength tunable CW diode seed laser, electro-optical modulator (EOM) and an Erbium doped fiber amplifier (EDFA) in a Master Oscillator Power Amplifier (MOPA) configuration. DAC depicts digital-to-analog converter, ADC analog-to-digital converter, DLEM range finder. The CO₂ cell serves as wavelength reference to calibrate the seed laser sweep current ramp. (b) Main data processing steps with real data sequences.

The signal strength of the light, backscattered by the topographic target, is recovered through a programmatically realized digital lock-in operation. As the seed laser wavelength is swept over 20 wavelengths a corresponding transmission

spectrum with 20 values is obtained. Stacking (averaging) subsequent spectra reduces random noise [Fig. 1(b)].

A model based on target range, air temperature, pressure and humidity computes a Lorentzian absorption line, which is fitted to the average spectrum in real time [Fig. 1(b)]. The measured spectrum acts as a fingerprint and, by means of an algorithm, allows automated rejection of data associated with interference related artifacts or other non-statistical noise sources. Linear and nonlinear baseline changes are accounted for in the fitting procedure via a background polynomial [24]. The differential optical depth (DOD) is deduced directly from the fit as $DOD = \frac{1}{2} \ln(\min[F])$, where F is the best fit transmittance, and is used to determine the CO₂ path density (in m²) or converted to path averaged mixing ratios (in ppm) as $X_{CO_2} = 10^6 DOD / (r \Delta \sigma N_{air})$, where r is the target range measured by the range finder (Fig. 1a), $\Delta \sigma$ the differential absorption cross section of CO₂ and N_{air} the air number density [21]. For each DOD value, the model covariance matrix scaled with the residual standard deviation of the fit (root mean square error) provides a meaningful confidence estimate.

For airborne use, drastic changes in reflectivity (e.g. water to rock) during a wavelength sweep should be minimized, i.e., the overlap of beams of different wavelengths maximized. At a representative altitude of 1600 m the beam diameter on the ground would be ~ 3 m. For a typical air to ground speed of 50 m/s this results in a maximum sweep time of the order of 50 ms (20 Hz rate). The default integration time was thus set to 1 s, resulting in a stack of 20 spectra per DOD value. The FPGA of the NI cRio controller was programmed such that a chunk of data [a section of which is depicted in Fig. 1(b)] was acquired uninterrupted before streamed to the PC where the data underwent real time processing and were saved for post-processing.

Fiber lasers offer excellent beam characteristics with nearly Gaussian beam profile, are alignment free and compact. Although that makes them ideal for the proposed instrument, their extremely narrow line width comes at the expense of a long coherence length (>2000 m at ~ 1 kHz line width), which aggravates susceptibility to interference related intensity noise during propagation in the atmosphere [25]. A tunable diode laser with line width ~ 200 kHz was therefore used as seed laser. The associated coherence length is ~ 200 m, while the associated error in DOD for the broader line width remained negligible ($<0.001\%$). At typical path lengths of 500 m to 2000 m this effectively suppresses noise due to air turbulence related interference.

To normalize optical power fluctuations a small fraction of the transmitted power had to be tapped off. Employing an integrating sphere with opened beam entrance and exit for that purpose solved the following issues:

- Wavelength dependent intensity changes occurring in the optical tap and the following optical elements (fiber, fiber/air interface, transmitting collimator).
- As the last optical element is the energy monitor, interference fringes and the associated non-stationary intensity noise was minimized.

Figure 2(a) shows a calibration test in the laboratory using a 10 cm long cell with pure CO₂ at atmospheric pressure. The topographic target was a wall at 7 m distance. The standard error of the time series corresponds to an uncertainty of $\sim 6\%$ of the CO₂ concentration in the cell. The sweep time was 500 ms. As indicated

by the Allen deviation in Fig. 2(b), averaging up to 9 s of data effectively reduces the error. Figure 2(c) shows *DOD* values measured outdoors for four different hard target ranges fairly following the expected linear trend [Fig. 2(d)].

Fig. 2. Performance of LARSS. (a) Stability test in the laboratory showing transmittance versus time through a 10 cm long cell containing 100% CO_2 . The laser beam traversed the cell once. (b) Allen deviation of (a). (c) Probing ambient CO_2 from the roof of the laboratory (star) at 4 different paths using house roofs as targets. (d) Resulting *DOD* (average of ~ 1 min of data each) versus range for the 4 paths in (c) with error bars (1 STD).

The instrument was transported to Java, Indonesia [Fig. 3(a)], on board a commercial passenger airliner and then mounted inside a small, single engine, propeller aircraft (Pilatus Porter PC-6). This airframe featured an open nadir viewing hatch. Telescope, laser transmitter and range finder (TX/RX unit) were mounted over this port [Fig. 3(b)]. During a relatively short flight on the 25 September 2016 the aircraft flew a landing approach near the city of Pangandaran [Fig. 3(a)] providing a dense variety of test scenarios for LARSS. Including measuring CO_2 from various altitudes (optical range) to a variety of surfaces and terrain including land, both agricultural and forest, ocean and small bodies of water. The corresponding *DOD* values are shown in Fig. 3(d). Given that this was a first and ad-hoc trial, the *DOD* satisfactorily follows the expected trend. Laser light scattered off cumulus clouds [Fig. 3 (c)] gave rise to low *DOD* in the first couple of minutes. The weather record for Pangandaran stated a 59% cloud cover (<https://www.worldweatheronline.com/pangandaran-weather-history/jakarta-raya/id.aspx>). The instrument used a 1 s integration with a corresponding ground track of ~ 60 m, thus some of the averaged spectra is composed of mixed optical paths from both surface and cloud returns. This leads to a reduced, effective absorption and causes the observed increase in scattering and lower *DOD* values. At altitudes below 800 m, 1 s worth of raw data featured strong fluctuations in return intensity, indicating changes in surface reflectivity manifesting in large deviations from the theoretical *DOD*. The time series of flight data shown in Fig. 3 starts over the ocean then at 13:08 transitions to land. Flying over optically heterogeneous terrain: sandy beaches, rice paddies and small standing water bodies as well as fields and forests in fast transition. Overland on approach the plane flew at only ~ 500 m altitude. The low altitude caused a 0.9 m beam diameter on the ground, while the plane was still traveling 2.9 m per acquired spectrum (per 50 ms). The consequential low overlap between

beam spots of different wavelengths caused reflectivity variations and thus considerable changes in signal strength during each wavelength sweep, hence scattered *DOD* values [Fig. 3(d)].

Fig. 3. Airborne operation of LARSS. (a) Map of Java and flight GPS track near Pangandaran. BK depicts Bledug Kuwu mud volcano. (b) LARSS mounted in the aircraft with main unit and TX/RX unit. (c) Photo taken during measurement flight looking south. (d) Differential optical depths along the flight path. Data from spectra rejected are not shown. Reasons for rejection include high background noise, low absorption and strong mechanical vibrations. (e) Associated X_{CO_2} .

Since the range finder was non-operational due to humidity related issues, GPS data along with profile information from Google Earth was used to reconstruct the flight altitude above ground and hence the target range. Those *DOD* undoubtedly not associated with cloud reflections and unbiased by reflectivity variations were converted to X_{CO_2} [Fig. 3(e)]. These values have an average uncertainty of 10% (ca. 40 ppm) and agree with the average of 390 ppm from a dispersive in situ IR spectrometer onboard the Pilatus, measured at altitudes between 1000 m and 2000 m.

A ground-based measurement at Bledug Kuwu (BK) mud volcano in Central Java [Fig. 4(a)] was carried out on 27 September 2016. The large volume of mud extruded by this mud volcano limited access to the main vent area, making it an ideal site for LARSS. Gas was ejected in the form of exploding bubbles, which lasted ~ 2 s. To resolve these events, the integration time was set to 0.5 s, corresponding to 10 stacked spectra per *DOD* value. The TX/RX unit was scanned across a gas plume emerging from the main vent. The optical power used was 0.7 W. So an eye safe power density (1000 W/m^2) was reached after 17 m distance from the transmitter. To assess feasibility the scan was carried out by hand [Fig. 4(a)]. Since the plume transport vector had no vertical component the scan was performed vertically from a tower 5.3 m above ground [Fig. 4(b)]. This would result in a profile of path averaged CO_2 concentrations perpendicular to the plume transport direction, spanning the whole diameter of the mud volcano of nearly 700 m. Figure 4(c) shows one of the resulting

profiles acquired by tilting the TX/RX unit from -4 degrees (-70 mrad) until close to 0. The tilt angle was measured with a tilt meter and fairly linear with time. The asymptotic error of the linear fit was 8% of the scanning rate of 0.6 mrad/s. The range per tilt angle was therefore reconstructed from the tower height and the tilt angle. X_{CO_2} were computed from the DOD and ranges. Values peak at 2400 ppm at 140 m distance. Highest CO_2 concentrations approximately cluster around the main vent. Bledug Kuwu encompasses also minor vents that contribute to the plume profile, explaining the peak at -45 mrad.

Fig 4. Ground based operation of LARSS. (a) Photo showing the TX/RX unit resting on the handrail of the tower aiming at the plume of Bledug Kuwu mud volcano, visible by condensed water vapor, also shown in the blow-up along with the vent. The scan aimed at the section downwind of the vent. (b) Sketch of the measurement geometry (not to scale). u depicts the plume transport vector. (c) DOD versus tilt angle and corresponding range along with associated X_{CO_2} with confidence interval (1 STD). Also shown is the ambient mixing ratio.

The X_{CO_2} confidence interval in Fig. 4(c) accounts for the signal-to-noise-ratio (random error) and range error and typically corresponds to 10%, mainly caused by the lack of the range finder (range uncertainty 20 m instead of 1 m causing 40% decrease in confidence). The rather short 0.5 s integration time further added to a dominating spectral fitting error. Rain during the measurement contributed to noisy spectra. In addition, for the longer ranges the scan angle increment $\Delta\beta$ produced a field of view, which included a wide topographic target range (~0.5 m near -70 mrad versus ~25 m near -10 mrad). This significantly altered the target reflectivity during each sweep, producing an additional interfering signal in the transmittance spectra. Nonetheless, the measurement demonstrated that large, vented and diffuse emissions may be probed even hand-held to attain gas profiles suitable for gas flux computation.

For a ground-based sight, with the range finder being operational and a 1 s integration time the uncertainty of the path length concentration product (detection limit) is currently ~9500 ppm.m.

In conclusion, using off-the-shelf technology as well as original data acquisition and processing a unique platform could be realized and demonstrated in very harsh environments. This instrument is seen as a versatile, easy to install geological field measurement tool that provides more representative data for remote monitoring of local CO_2 emissions for both Earth and environmental science. The system has been performing a first-time measurement of CO_2 concentration profiles at the Bledug

Kuwu mud-volcano, which, to our knowledge, is the first single-ended remote sensing of degassing at a mud volcano. Further refinements are needed to improve measurement precision, in particular for the much more challenging airborne acquisitions, and for turnkey operation. The sweep rate will be increased, making the system more robust against density fluctuations in the atmosphere as well as minimizing target reflectivity variations during sweeping and allow a higher number of spectra to be averaged per integration time.

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