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Application of Monitoring Methods for Remote Detection of Atmospheric CO_2 - Concentration Levels during a Back-Production Test at the Ketzin Pilot Site

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Abstract

Reliable detection and assessment of near-surface CO_2 leakages from storage formations requires the integration of various atmospheric monitoring techniques. In October 2014 at the Ketzin pilot site, formerly injected CO_2 was retrieved from the reservoir and vented to the atmosphere ("back-production experiment"). This experiment was accompanied by atmospheric monitoring methods applying meteorological approaches and ground-based optical remote sensing techniques. The main aims of the atmospheric monitoring were: a) observation of gas dispersion in lower atmosphere, b) determination of maximum CO_2 concentration and c) identification of the main challenges associated with the monitoring using field set up under typical environmental conditions.

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Keywords: Ketzin pilot site, monitoring CO_2 releases, atmospheric monitoring, open-path Fourier-transform infrared spectroscopy

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1. Motivation

The detection and monitoring of areas susceptible to potential CO_2 release plays a fundamental role within a hierarchical monitoring concept during CO_2 storage projects. Using such a concept, atmospheric monitoring methods allow for scientifically monitoring areas on larger scales to identify zones with increased atmospheric CO_2 concentrations. This in turn can help identify target areas for subsequent detailed investigations. Ground-based atmospheric measurement methods can be used to monitor the atmospheric composition of the lower atmospheric boundary layer and therefore can be important tools to help control greenhouse gas emissions [1, 2, 3].

The integrative hierarchical monitoring concept applied within the framework of the MONACO project (MONACO = "Monitoring approach for geological CO_2 storage sites using a hierarchical observation concept") showed that the use of remote sensing techniques (e.g., open-path Fourier-transform infrared (OP FTIR) spectroscopy, Eddy Covariance (EC) measurements) combined with meso-scale measurements (geophysics and chamber-based soil CO_2 flux measurements) and local in-situ measurements (Direct Push Technology) enables a more reliable characterization of storage sites for the further validation of potential risks [4]. Especially at larger scales, the application of ground-based optical remote sensing methods (such as OP FTIR) is a trustworthy atmospheric monitoring technology which can be used to detect the concentrations of various gases in ambient air - in real-time and over large distances [5].

The atmospheric monitoring methods were applied at the Ketzin pilot site for geological CO_2 storage during a field experiment in October 2014 within the project COMPLETE (CO_2 post-injection monitoring and post-closure phase at the Ketzin pilot site). This field experiment ("back-production experiment") retrieved formerly injected CO_2 from the reservoir via one well and released it to the atmosphere via a vent stack. Applied monitoring methods were comprised of an integrative system using point sensors to observe near-surface CO_2 concentration, micrometeorological approaches using Eddy Covariance equipment and ground-based optical remote sensing techniques based on open-path Fourier-transform infrared spectroscopy. The main aims of our atmospheric monitoring were: a) observation of gas dispersion in the lower atmosphere, b) determination of maximum CO_2 concentration values and c) identification of the main challenges associated with monitoring point source leakages using the proposed methodological set up under typical environmental conditions.

2. Test site

The ground-based atmospheric monitoring tools were installed at the Ketzin pilot site, the first European onshore storage pilot site for the geological storage of CO_2 in the German Federal State of Brandenburg, about 25 km West of Berlin. This multidisciplinary research project was established in 2004 to advance the scientific understanding of CO_2 storage and its related processes (those caused by CO_2 injection and migration), and to develop successful communication and public outreach strategies. The results from the Ketzin project demonstrate the feasibility of geological CO_2 storage on pilot scale [6]. Between June 2008 and August 2013, a total amount of 67 kt of CO_2 was injected into the saline aquifer sandstone formation with an injection depth of 630 – 650 m. By the end of the injection period, the Ketzin pilot site entered into the post-closure phase [7].

The back-production test (as part of the post-closure experiments undertaken) was performed between October 15th and 27th 2014 and a total amount of 240 tons of CO_2 and 55 m^3 of brine were safely back-produced via a former injection well in the CO_2 storage reservoir. Various re-production rates (maximum rates up to 3,600 kg/h during start-up, rate 800 kg/h from 15th to 20th and 23th to 27th, rate 1,600 kg/h from 20th to 22nd [8]) were realized over the two-week period to validate technical feasibility. This experiment provided data on the pressure and temperature evolution during the back-production of CO_2 from the storage formation into the ambient atmosphere, data on the chemical composition of the produced brine and CO_2 as well as data on the behavior of atmospheric CO_2 concentrations. A technical description of the test and the results besides the atmospheric monitoring are described in [9].

3. Atmospheric monitoring methods

During the back-production test, open-path Fourier-transform infrared (OP-FTIR) spectroscopy, the Eddy Covariance (EC) method, and distributed CO_2 point sensors (based on non-dispersive infrared NDIR detectors in combination with on-site meteorological parameter acquisition systems) were applied to identify temporal and spatial variations in atmospheric CO_2 concentrations in real time. This paper focuses on the advantages and limitations of passive OP FTIR spectroscopy in such an integral monitoring frame work.

Open-path spectroscopy is a method which is suited for detecting various chemical compounds simultaneously in ambient air, in order to remotely monitor volatile emissions in real-time and to investigate larger sampling areas. Infrared spectroscopy is based on the effects of molecular vibrations caused by interactions between infrared (IR) energy and molecules. During OP FTIR measurement, IR energy is transmitted through the open, unobstructed atmosphere from an IR source (natural or artificial) to the detector device. Measurement of the absorption loss along an optical path in ambient air represents the ‘path integrated gas concentration value’ (PIC) for the whole optical ray path.

Most atmospheric target gases (e.g., CO_2, CH_4, N_2O) display distinct absorbance or emission patterns in IR spectra, super-imposing the blackbody radiance. The identification of gases is based on selected absorption bands in significant spectral areas. The wave number range considered is between 700–4000 cm^{-1} (LWIR wave length 2-14 μm). Open-path spectroscopy can be applied in two measuring modes. Natural IR background radiation is utilized for the passive measurement mode, while an artificial broadband IR source is required for the active measurement mode. The maximum path length is up to 3,000 m for the passive mode and up to 300 m for the active mode. In this paper, we examine the results of OP FTIR spectroscopy in the passive mode.

4. OP FTIR spectroscopy field set-up

During the field test, the active OP-FTIR system, passive OP-FTIR spectrometer and one of the EC monitoring stations were installed at distances approximately 2 m apart from each other. All of these devices were placed to the east of the vent stack at a distance of 110 m away from the vent stack (Fig. 1).

Passive OP-FTIR spectroscopy is based on the spectral analysis of ambient IR radiation in the 700 – 1300 cm^{-1} wave number range, and is limited by CO_2 and H_2O total absorption regions. Numerous chemical compounds have a typical IR signature in this fingerprint region, where no artificial IR source is needed. To collect these passive spectra, a BRUKER RAPID (Remote Air Pollution Infrared Detector) device was used, which is based on a mercury cadmium telluride (MCT) IR detector. The instrument is a flexible and mobile system for the analysis of atmospheric trace gases and air pollutants at distances of up to 3,000 meters. Passive OP FTIR spectroscopy was applied in imaging mode for the determination of the spatial distribution of CO_2 concentrations in the vicinity of the vent stack. Passive scans were carried out several times after start of the experiment by 25 single passive OP-FTIR measurements in 22 different view directions in an area at an angle of 80° around the vent stack.

Furthermore, continuous monitoring of a single optical path direction (straight toward the vent-stack, seen in Fig.1) was conducted to observe any temporal CO_2 variations; measurements started already on 13th October and ended on 23rd October 2014. The passive optical path was chosen alongside the active optical path to observe any temporal variations. As such, passive single beam spectra were achieved in a 12 minute sampling interval for the whole experiment – 1,008 spectra in total. Additionally, meteorological data were required for OP FTIR data interpretation. These parameters (such as air temperature, wind speed and wind direction) were acquired from the EC station near the IR detectors. During the experimental phase, the prevailing winds came from the west (Fig. 1). However, periods with winds coming from the south represent adverse conditions for the detection of CO_2 along the optical pathway.

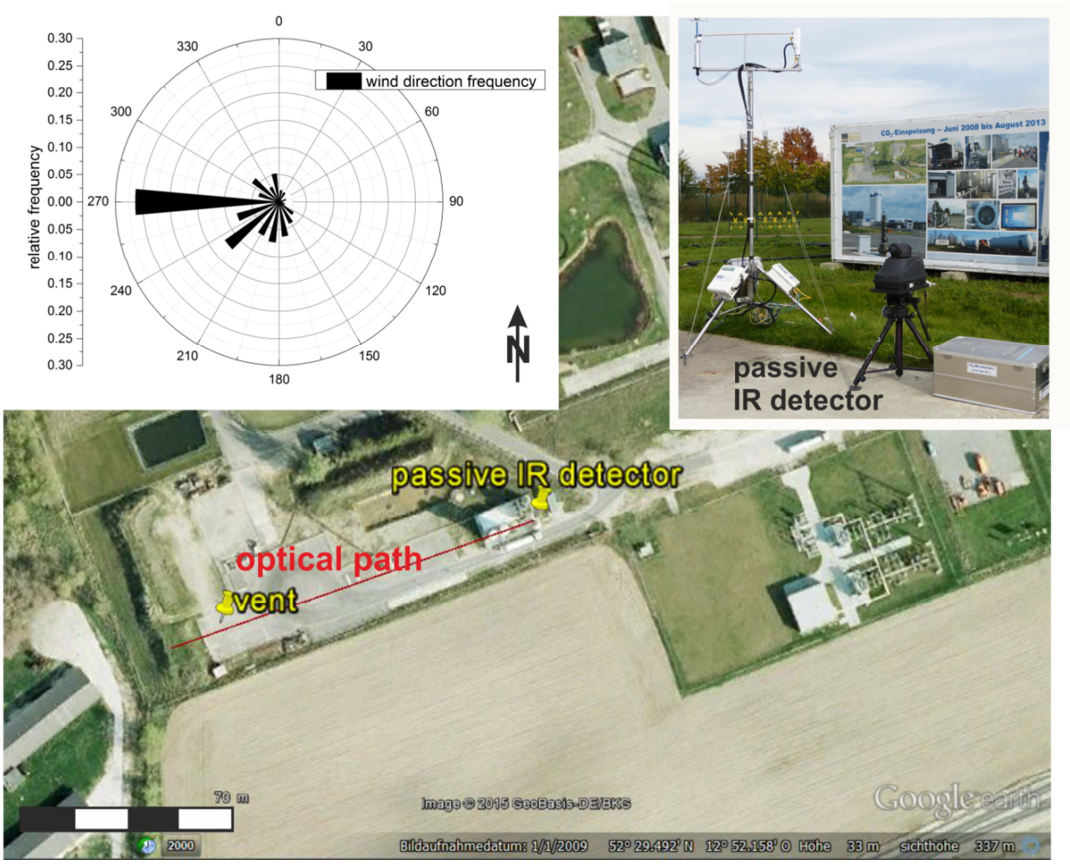

Fig. 1. Instrumental set-up at the Ketzin pilot site for atmospheric monitoring. Passive detector was installed at a distance of 110 m east from the vent stack. Meteorological data were acquired using eddy covariance instrumentation next to the passive IR detector. During the back-production phase, the prevailing winds came from Western direction.

5. Results of passive OP-FTIR spectroscopy

The data processing of the passive spectra includes the calculation of brightness temperature spectra from measured single beam spectra, the analysis of temperature information for background and ambient air conditions, and the analysis of signal behaviour at the significant CO_2 absorption wavenumber 720 cm^{-1}. The brightness temperature spectra reveal distinct variations in CO_2 absorption depending on the direction of the optical path and the time of measurement.

The first experimental part was conducted to obtain information about the spatial distribution of increased CO_2 concentrations in the vicinity of the vent stack. Figure 2 shows parts of the brightness temperature spectra in the 720 cm^{-1} wavenumber region. Obviously, measurements taken near the vent stack are subject to increased amplitudes in the CO_2 absorption band. Furthermore, we observed an increase in determined brightness temperature, which is an indicator for increased atmospheric temperature along the optical pathway. The recognized anomalies in CO_2 absorption and in temperature are caused by the vented gas, which consists of more than 97 % CO_2 [10], is produced at a rate of ~ 800 kg/h from the storage formation, and was heated to 40 – 50 °C prior to venting to avoid any dry-ice formation.

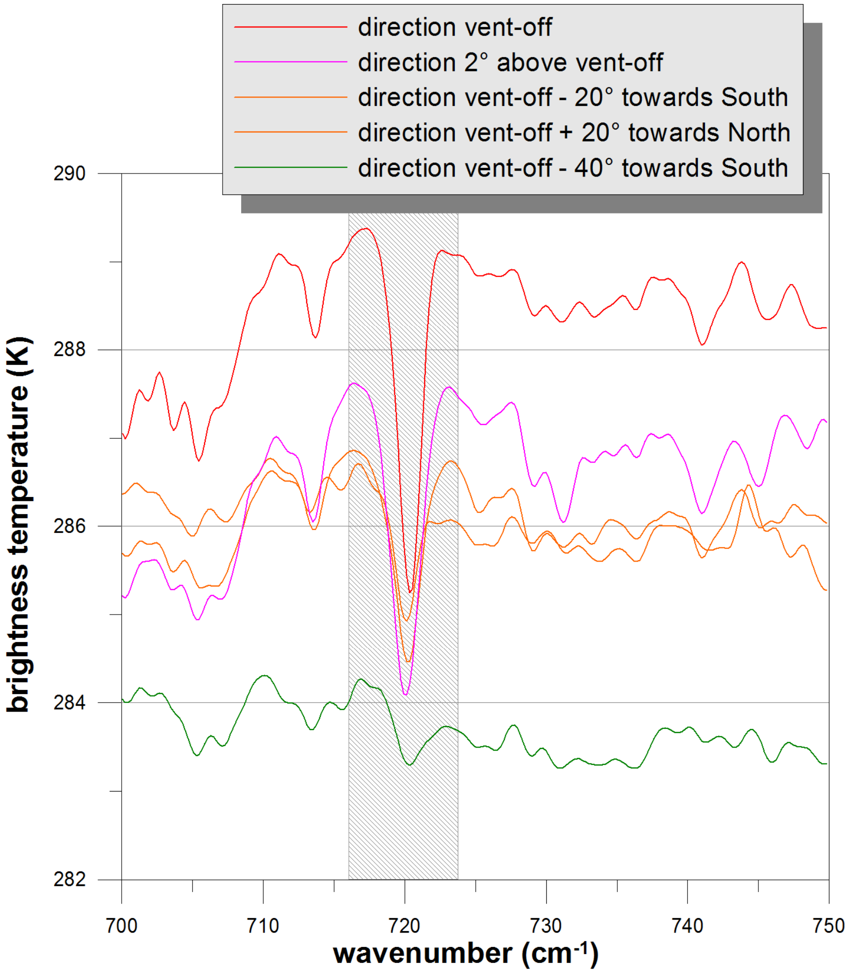

Fig. 2. Section of brightness temperature spectra measured in different directions near the vent stack. There are distinct CO_2 signatures in the spectra at the 720 cm^{-1} absorption band, especially for the measurements in closer proximity to the stack. Obviously, an increased brightness temperature around the stack can also be observed in the spectra, which is caused by the higher temperature of the back-produced gas.

In addition, the results indicate an observable rise in relative change of atmospheric CO_2 concentration after the start of back-production in relation to baseline conditions. Based on results from the scanning technology, all measurements taken in different optical path directions were compiled to form an image illustrating the spatial distribution of relative changes in CO_2 concentration related to baseline conditions (Fig. 3). The data shown here were acquired 90 minutes after the start of CO_2 venting. Both parameters, relative CO_2 concentration and temperature, display distinct anomalies near the vent stack due to the occurrence of vented gas in the atmosphere. Furthermore, a secondary anomaly zone can be distinguished in both parameters at the right edge of the scanned image. The technical facilities for the experiment were located at this position (including a diesel operated heater). These installations are likely to be the cause of the observed secondary CO_2 and temperature anomalies.

The second experimental task comprised the determination of temporal changes in relative CO_2 concentration in the vicinity of the vent stack. These were obtained with passive OP FTIR spectroscopy to derive information on influencing parameters and establish any detection limitations of CO_2 releases from a point source. This information is relevant for the industrial implementation of atmospheric monitoring using optical remote sensing techniques such as OP FTIR spectroscopy, especially in the case of potential leakages that occur as a result of improperly abandoned wells.

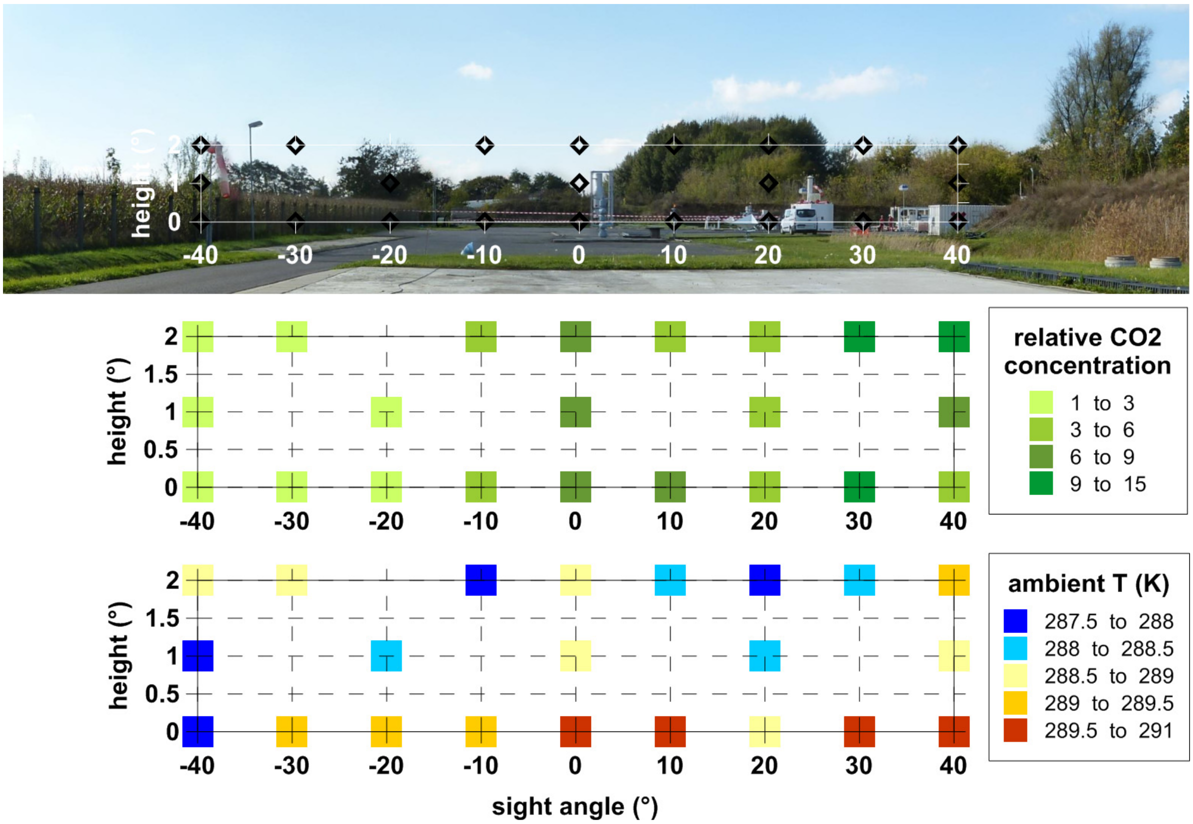

Fig. 3. Imaging of ambient air conditions using passive OP-FTIR scanning technology.
Top: Photo shows the detector field of vision and the directions of the optical pathways.
Middle: 90 minutes after the start of back-production, a distinct increase in CO_2 concentration relative to the baseline conditions is determined. There are two zones with raised CO_2 concentrations. The primary CO_2 source is at the vent stack, which is located at sight angle = 3°. A secondary relevant CO_2 source is detected at sight angle = 30°. This source might be caused by the diesel engine of the back-production facility.
Bottom: There are also two zones with increased ambient temperatures which correlated to the CO_2 anomalies.

Figure 4 displays the time series of temporal variation of relative CO_2 concentration change relative to the baseline derived from passive OP-FTIR spectra. This is compared to wind conditions during the back-production experiment. The experiment started on the 15th October at noon while highest back-production rates occurred, including maximum values of 3.600 kg/h over a period of one hour. In the timespan from October, 15th to 20th, the mean back-production rate was approximately 800 kg/h and then was taken up to 1.600 kg/h until October, 22nd, where production was stopped in the afternoon prior to night hours. The determined relative changes of CO_2 concentration values seem to be very noisy and vary over a wide range during the experiment. However, the increased back-production rate in the beginning of the experiment is clearly detected. Nevertheless, the observed CO_2 concentration variations need to be jointly considered with the meteorological data. Obviously, there is a correlation between increased relative CO_2 concentration fluctuations and wind conditions. In particular, situations with wind coming from the Western direction are responsible for transporting the CO_2 cloud from the vent stack into the optical pathway. Wind from the South avoids being as easily detectable as the released CO_2. However, in calm wind situations, the accumulation of CO_2 can be observed while refreshing winds into the direction of the optical path after these calm conditions caused raised CO_2 signals.

6. Conclusions

The applicability of passive ground-based OP FTIR spectroscopy was validated as a possible method for continuously monitoring the fluctuations of atmospheric composition, in terms of identifying areas with increased gas concentration. Applying the technique during the back-production experiment at the Ketzin pilot site allowed to obtain data from an artificial analogue to a leaking well and to test the applied methodology, concerning its ability to reliably detect such kinds of point sources. With our measurements, we could show that temporal and spatial variability in atmospheric CO_2 concentration was traceable with the chosen experimental set-up. With the scanned image, it was possible to distinguish between two emission sources. The temporal observations outlined the huge variability in the measured relative changes in CO_2 concentration and emphasised the necessity to measure baseline concentrations, so that natural CO_2 diurnal variability as well as any additional meteorological parameters can be registered. This helps to gain a better understanding of dilution and atmospheric transport processes.

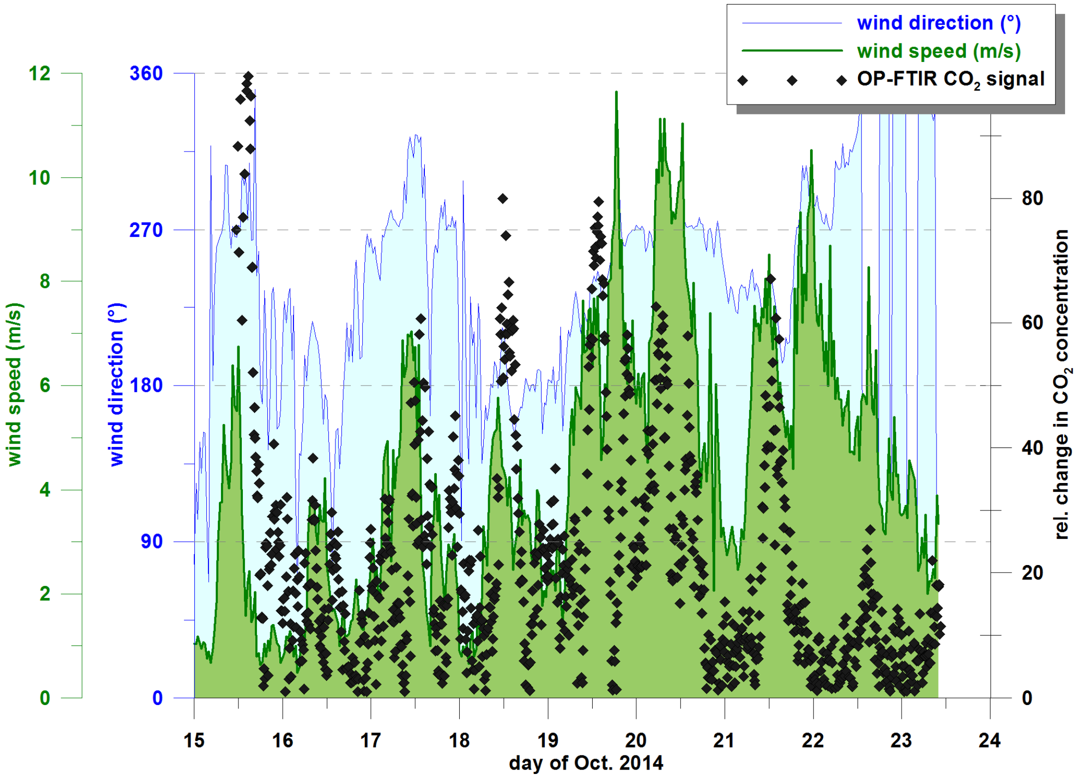

Fig. 4. Temporal variation of relative change of CO_2 concentration relative to baseline conditions derived from passive OP-FTIR spectra compared to wind conditions (wind speed, wind direction) during the back-production experiment. The wind conditions are the primary influencing parameters on the detectability of the CO_2 cloud in the atmosphere. Wind from the Southern direction reduces detectability, while the source is located North of the optical path. Furthermore, calm wind situations are responsible for accumulation processes and increased amplitudes were simultaneously observed with stronger winds.

The data clearly demonstrate that there is a strong dependency on meteorological parameters (wind direction, wind speed). Windy situations favour atmospheric mixing processes and cause a rapid dilution of the vented CO_2. Therefore, meteorological conditions have to be taken into account for the interpretation of atmospheric monitoring and the determination of detection thresholds for further industrial automation. Increased atmospheric CO_2 concentrations could be reliably detected with the applied combination of monitoring methods during calm weather conditions. In contrast, windy situations can cause a rapid dilution of the vented CO_2, resulting in no increased atmospheric CO_2 concentrations levels determined with the applied remote sensing methods - even in the vicinity of the CO_2 vent stack. Adverse wind directions are especially important, as affecting the direction of the optical path can result in obtaining non-interpretable data.

OP FTIR spectroscopy has been validated in this study as suitable monitoring technique. It is an appropriate optical remote sensing technique working in the lower atmospheric boundary layer on larger scales. Our results demonstrate, however, the application of OP FTIR spectroscopy alone cannot cover all atmospheric monitoring aspects of industrial CO_2 storage operations and must be used as an integral part of a comprehensive monitoring concept, consisting of methods that focus on numerous environmental parameters at different temporal and spatial scales.

Nevertheless, the experimental results provide the information required to use the method as part of a further industrial set-up of optical remote sensing techniques in order to monitor atmospheric quality at potential geological CO_2 storage sites. Thus, for a reliable interpretation of OP FTIR spectroscopy data, all known CO_2 sources need to be characterized (natural emissions, artificial emissions and industrial sources) and measured in terms of baseline monitoring to quantify their input. The main advantage of OP FTIR spectroscopy is its ability to simultaneously verify other target gases such as CH_4 with one single measurement. This feature can be used to characterize and distinguish between different emission sources. In addition, facility infrastructure and prevailing wind conditions need to be assessed and must be taken into account when implementing such a monitoring program.

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