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Testing a simple and low-cost method for long-term (baseline) CO₂ monitoring in the shallow subsurface

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Abstract

Implementation of geological CO₂ storage requires monitoring for potential leakage, with an essential part being establishment of baseline CO₂ in soil gas. CO₂ concentrations and weather parameters were monitored for ~2 years at three locations in the Netherlands. CO₂ concentrations in soil ranged from 0.1 to 28% and were variable with site, depth and soil type. Statistical models (based on weather parameters) corresponded well with the measured CO₂ time-series, having best fit with soil temperature. This method can detect large CO₂ anomalies, however not smaller leakages. The monitoring method is robust and low-maintenance, making it a relatively low-cost surveillance possibility.

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1. Introduction

A major obstacle for geological storage of CO₂ is the potential leakage of CO₂ towards the surface. Monitoring in the deep subsurface by e.g. geophysical measurements, can be used as an ‘early-warning-system’ in order to commence with leakage mitigation. Only if leakage from the storage site *would* occur and the applied early-warning-

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system and mitigation techniques are insufficient could CO₂ seepage towards the surface occur. Potential ‘risky’ pathways are wells, caprocks, spill points, faults and fractures (Rütters et al., 2013). Finally, the CO₂ could reach the vadoze zone of the shallow subsurface before it escapes into the atmosphere. Potential leakage would occur gradually, thereby slowly increasing the natural CO₂ values in the soil gas. Yet, monitoring of the gaseous CO₂ concentrations and fluxes in the shallow subsurface could serve several purposes. First of all, it would reassure the public that abnormally high CO₂ concentrations will be detected well before any safety limits are exceeded. Secondly, if abnormally high concentrations are detected in the shallow subsurface, remediation measures would be taken immediately to prevent environmental damage. Consequently, measurement of leaked CO₂ fluxes can serve a penalty system in the EU Emission Trading System (ETS) (Rütters et al., 2013).

Because natural CO₂ concentrations in the vadoze zone are variable, both in space and time, future monitoring depends on a site-specific, comparative dataset obtained prior to CO₂ injection. This dataset would serve as a baseline. To comprehend the seasonal variation of CO₂ concentrations in the vadose zone, it is necessary to measure background concentrations for at least one year (Kronimus & Starec, 2008). The concentrations can then be used for predictive modelling of natural CO₂ concentrations with time (e.g. Kronimus & Starec, 2008). The focus of this study is to develop and test a simple, low cost and long-term monitoring system for the Dutch shallow subsurface.

1.1. Background

Natural carbon dioxide concentrations in the vadoze zone are affected by microbial activity, respiration of root systems, photosynthesis, leaf respiration, and environmental factors such as soil and air temperature, soil moisture, wind speed and barometric pressure (e.g. Boone et al, 1999; Curiel Yuste et al, 2007; Bekele et. al, 2007; Cable et al, 2013; Singh et al., 2009; Lewicky et al., 2010; Schoelmer et al., 2014). Spatial and temporal variability in the shallow subsurface CO₂ concentrations represents differences in rates of CO₂ production and transport caused by the complex interactions between these biotic and environmental factors. A positive relation was identified between soil temperature and CO₂ concentration because increasing temperatures give rise to root respiration and oxidation of soil organic matter (Boone et al, 1999; van Eijndhoven 2005; Starec, 2006; Curiel Yuste et al, 2007; Cable et al, 2013), and methanotrophic respiration (Starec, 2006). Similarly, soil moisture enhances microbial and metabolic activity as well as oxidation of organic matter, thus increasing CO₂ production. However, high water content saturates pore spaces with water, which results in the opposite effect (e.g. Jassal et al., 2004). At high barometric pressure atmospheric CO₂ can migrate into the soil (e.g. Schloemer et al., 2014). Hinkle (1994) postulated that the decrease of barometric pressure eases the escape of CO₂ from the soil. Non-weather parameters that are of influence on the CO₂ concentrations in the shallow subsurface are soil type, land use (Curiel Yuste et al, 2007; Boone et al, 1998; Cable et al, 2013; Singh et al., 2009) and pH (Kharaka et al., 2006; Singh et al, 2009).

This work builds on previous shallow monitoring studies at TNO by Starec (2006) and Kronimus & Starec (2008). It was noted that measurements in open tube sensors were hampered by influence of barometric pressure and wind intensity (van Eijndhoven, 2005; Starec, 2005; Kronimus & Starec, 2008). Better results were achieved when sensors were buried into the soil and backfilled with the previously dug out soil (Bekele et al., 2007).

2. Methods

2.1. Monitoring setup

Three different locations in the Netherlands with three different soil types were selected for this study (Fig. 1). These locations were selected mainly due their variability in soil type. The availability of power and internet, safety and an undisturbed environment also played a role in the selection. The Cabauw site southwest of Utrecht has a clayey peat soil and was monitored for 80 weeks using eight CO₂ sensors. The Uithof in Utrecht has predominantly sandy soil with a few silty clay beds. The monitoring continued for 100 weeks at this site. The soil at Lutjewad in Hornhuizen is mostly clay with a few sand beds and monitoring was undertaken for 60 weeks. Two CO₂ sensors were installed and monitored at Uithof and at Lutjewad. The soil at all locations is covered with shallow root vegetation and is not artificially fertilised.



Fig. 1. Map of the Netherlands with monitoring locations.

At each location CO₂ sensors (CARBOCAP® GMM221 carbon dioxide sensors from Vaisala) were buried into the soil at depths of 50 cm and 100 cm along with sensors for soil temperature and soil moisture (EKOPOWER). The CO₂ sensors measure the concentration of CO₂ in the soil gas and cover the concentration range of 0 to 20% (analytical error = 0.02sd). They self-calibrate to adjust to temperature and pressure changes. The sensors were connected to a weather station (Wireless Vantage Pro2) that was placed at all of the locations to monitor air temperature, barometric pressure, rainfall and wind intensity. The sensors and the weather station were connected to a computer station, which transmitted real-time data via internet to a TNO website. The costs for entire monitoring setup at Cabauw (8 sensors, weather station, IT and supplementary equipment) amounted to ten thousand euros.

2.2. Data analysis and predictive model

The data was recorded in irregular time intervals due to the self-calibration of the sensors. Consequently, the data was averaged to weekly time intervals with Matlab®, to create a smooth time-series for each site. The data was then compiled in sequence plots to provide an overview of the behaviour over time. The model approach was adapted from Kronimus & Starec (2008). Frequency analysis was performed to identify cycles and/or repetitive patterns. Coefficients of the linear equation that could best predict the CO₂ concentration were determined using one or more independent variables (in this case soil temperature, barometric pressure and soil moisture). Bivariate correlation (Pearson correlation) with SPSS was used to establish the a best fit for each weather parameter. The predictive model was developed using a simple linear regression model in SPSS Modeler (IBM, 2011). The models were developed based on the measurement data of the first half of the dataset. Subsequently they were applied to predict CO₂ concentrations based on the soil and weather conditions during the second half of monitoring period and later compared to the measured CO₂ concentrations.

3. Results

After the initial setup the monitoring systems did not require any further maintenance.

CO₂ concentrations are site *and* location specific. At the Uithof and Lutjewad locations groundwater was below the sampling depth of 1 m during the whole period of monitoring. The measured CO₂ concentrations fluctuated between 0.2 and 3.3, and 0.1 and 5.0 %, respectively. The highest and most variable concentrations were recorded at Cabauw, ranging from 0.3 to 28.3 %. Significant differences of up to 20% CO₂ were recorded from sensors located within a distance of a few meters (Fig. 2) and up to 12% with depth. Concentrations of above 20% must be treated with caution because the applied sensors are calibrated only up to 20% CO₂. At Cabauw, groundwater level

fluctuated between 0.5 and 1 m. CO₂ fluctuated seasonally at all sites with highest concentrations in the late summer period. A time lag of a few weeks was observed between the weather conditions and the changes in CO₂ concentrations, which is likely due to the delayed response of biological activity to changing weather conditions.

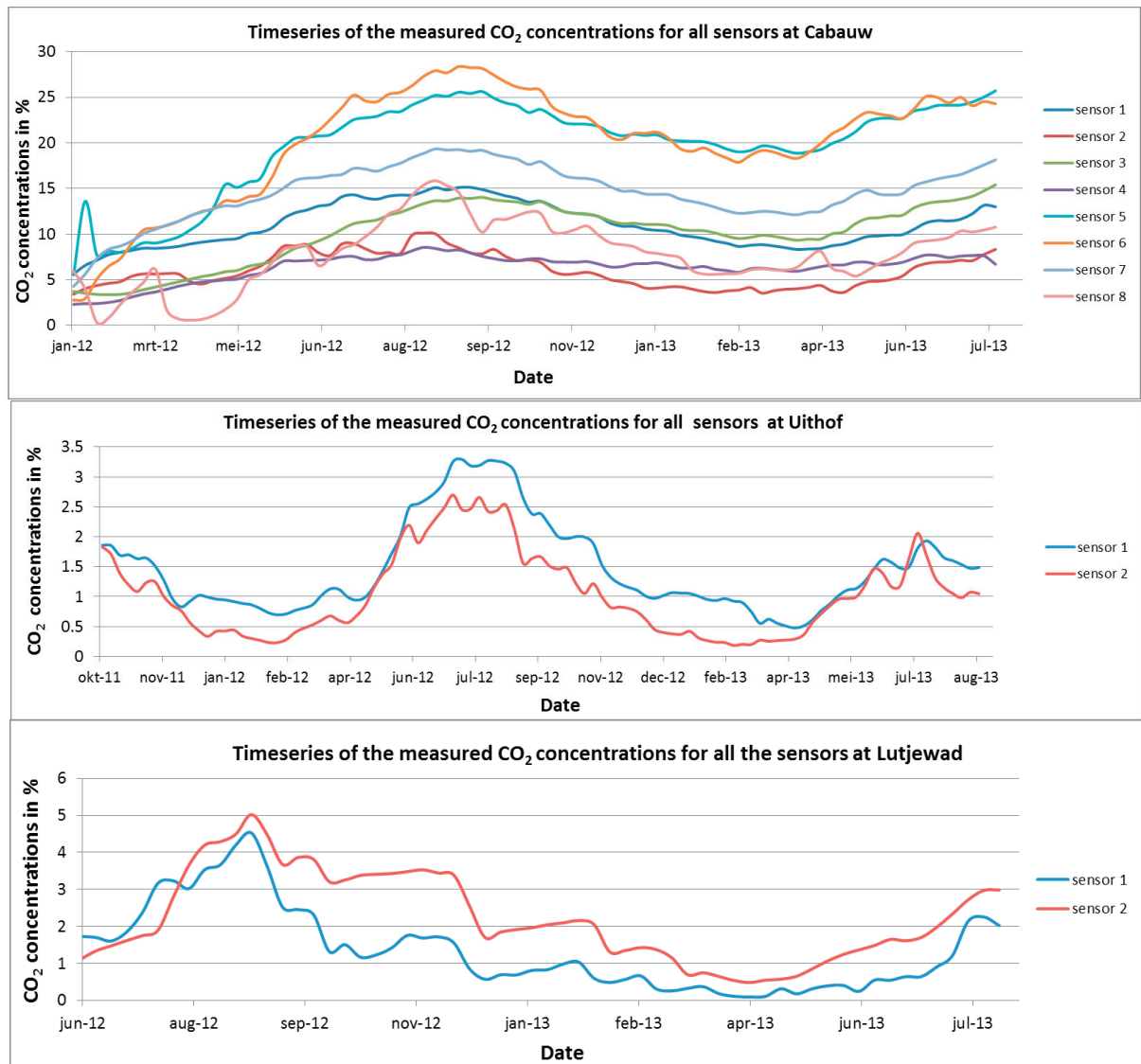


Fig. 2. Time series of CO₂ concentrations measured at the Cabauw, Uithof, and Lutjewad monitoring sites.

The measured CO₂ concentrations are best explained by the fluctuations in soil temperature, whereas soil moisture and barometric pressure showed a smaller effect. The best fit modelled CO₂ concentrations correlate generally well with the measured CO₂ concentrations (Table 1). The difference between the measured CO₂ and modelled (best fit) concentration was mainly between 0 and 3%, except for the location of Cabauw where the difference was occasionally as high as 10%.

Table 1. Overview of best-fit regression models. The modelled data is based either on temperature, soil moisture, barometric pressure or a combination of weather parameters from the first monitoring year and includes a lag shift, where applicable. *Correlation factors (R) between the fitted CO₂ concentrations and the measured CO₂ for the first half of the timeseries. **Modelled CO₂ concentrations compared to CO₂ measurements for the second half of the time series. An example of a table.

Sensor, depth	Model based on	Lag time	R	R
Location Cabauw		correction	year 1*	year 2**
Sensor 1, 1m	temperature + pressure	5 weeks	0.95	0.93
	temperature	5 weeks	0.97	0.95
Sensor 2, 0.5m	temperature	1 week	0.93	0.95
Sensor 3, 1m	temperature + pressure	7 weeks	0.91	0.65
	temperature	6 weeks	0.90	0.85
Sensor 4, 0.5m	temperature + pressure	5 weeks	0.91	0.77
	temperature	6 weeks	0.86	0.81
Sensor 5, 1m	temperature + pressure	6 weeks	0.92	0.76
	temperature	4 weeks	0.90	0.86
Sensor 6, 0.5m	temperature + pressure	6 weeks	0.93	0.70
	temperature	6 weeks	0.93	0.69
Sensor 7, 1m	temperature + pressure	1 week	0.92	0.91
	temperature	1 weeks	0.96	0.93
Sensor 8, 0.5m	temperature	7 weeks	0.87	0.81
Location Uithof				
Sensor 1, 1m	temperature	none	0.93	0.84
Sensor 2, 0.5m	temperature	none	0.95	0.89
	temperature + humidity	none	0.99	0.89
Location Lutjewad				
Sensor 1, 1m	temperature	none	0.75	0.78
	temperature + humidity	none	0.75	0.81
Sensor 2, 0.5m	temperature	3 weeks	0.70	0.82

4. Discussion

Seasonal variations on CO₂ are connected to more favourable conditions for root respiration in the summer months. High CO₂ concentrations at Cabauw (up to 28%) may be related to enhanced oxidation of the high amount of organic matter in peat and higher biological respiration due to the higher soil moisture. In addition the low permeability clay layer above the peat acts as a seal and prevents quick CO₂ escape to the surface (e.g. Alm et al., 2007; Brocca et al., 2007; Lewicki et al., 2010; Beaubien et al., 2013). Peat soil is not present within the top sediment at the other two sites. Furthermore, the top 1.5 m of soil at the Uithof location consist of sand, which is highly permeable and would thus allow CO₂ gas escape.

Previous studies observed CO₂ variation with soil/air temperature, soil moisture, barometric pressure and wind intensity (e.g. Curiel Yuste et al., 2007; Lewicky et al., 2010). In the present study best correlation was achieved between monitored CO₂ and soil/air temperature, which can be related to preferred plant respiration at optimal temperature conditions. Soil moisture, barometric pressure and wind intensity had a less significant effect on CO₂. The reason for this and also for the insignificance of pressure and wind intensity on the CO₂ concentration may be related to the depth of the sensors. The largest effect of soil moisture on respiration was found to be between 0 and 40cm (Bowden et al., 1993).

For the Cabauw location it is debatable if the measurements at a depth of 1 meter are eligible for analysis because of the height of the groundwater levels. Groundwater levels are above 1 meter but below 50 cm for the largest part of the year. The employed CO₂ sensors were not designed for measuring CO₂ concentrations in water, however

measured CO₂ concentrations could still be well reproduced by the models at 1m depth. Nevertheless, further verification of the fidelity of these sensors is needed.

We were able to develop a calibrated model which allowed to reproduce the measured concentrations with a maximum error of 3% for soils low in peat (Tab. 1, Fig. 3). Although the net difference between the measured and the modelled CO₂ concentration was partially larger at the peat-soil location of Cabauw, the correlation coefficient of the two datasets was generally higher. At the studied sites, significant CO₂ anomalies can be reliably detected with this method, thereby successfully serving the purpose of providing a safety feeling for the public as well as the ability to act as a warning system to prevent environmental damage. Small, continuous leakage will likely not be apparent without further analysis. Hence, for the purpose of a penalty system in the ETS only significant leakage can be incorporated using this technique.

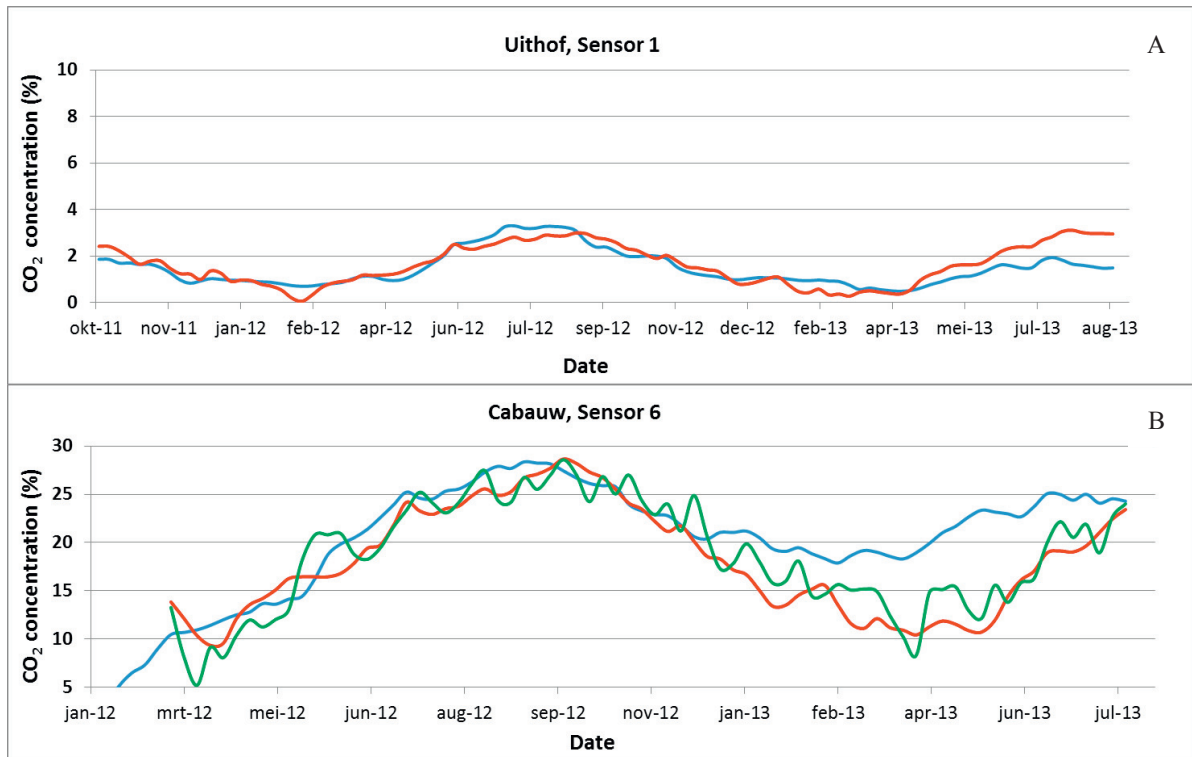


Fig. 3. Examples of CO₂ time-series (blue) and modelled CO₂ concentrations (red and green). A: example of a good model fit at Uithof site. B: Example of lower correspondence between modelled and measured CO₂ at the Cabauw site (note difference in scale). The red lines represent models based on soil temperature only, the green line includes the effect of pressure.

The sensors and the monitoring set-up functionality was robust throughout the monitoring periods. No further maintenance was needed after initial setup and the chosen sensors require calibration only once every two years. We are thus looking at a robust and relatively inexpensive method for monitoring in the shallow subsurface at a potential leakage site for detection of potentially harmful CO₂ concentrations. The disadvantage of the method is that it is spatially restricted and hence, selection of potential leakage sites is a crucial step prior to installation of the monitoring set-up. In addition, it should be noted that the detection of anomalies in areas with naturally high and variable CO₂ concentrations (like Cabauw) is less reliable than in areas with low natural concentrations. The assurance of the CO₂ measurement fidelity for the chosen sensors would benefit from testing with alternative sensors. Furthermore, a process based approach to clarify the source of CO₂ (whether shallow biological production or deep CO₂ source) would further strengthen the monitoring procedure. These should be the next steps towards a complete evaluation of the shallow CO₂ monitoring procedure.

Last but not least, a significant advantage of the applied monitoring procedure is the possibility of real-time publication of the data online. A website can be made public, allowing any interested party to view the data. A warning system can be installed when anomalies are measured. The increased transparency would enhance public acceptance towards onshore CO₂ storage.

4. Conclusions

We developed a monitoring system for baseline CO₂ concentrations in soil gas with the aim to detect significant anomalies resulting from possible CO₂ leakage from a storage reservoir. The set-up was tested at three different sites in the Netherlands. We observed significant differences in natural CO₂ concentrations between different sites as well as with depth. CO₂ concentrations are highly dependent on the soil type. The soil temperature had the highest effect on CO₂ fluctuations due to increased biological activity in the soil when temperature rises. The modelled CO₂ concentrations correlate well with measured CO₂ time-series. The difference between measured and modelled CO₂ concentrations ranges mostly between 0 and 3%.

Overall, this system is capable of identifying large deviations in CO₂ concentrations when deployed at a real CO₂ storage location, however small, continuous leakages will not be detected with this method. This is particularly the case for locations with naturally high background CO₂ concentrations. The monitoring system has been up and running for several years and has shown to be robust without much need for maintenance, providing a cost-effective surveillance opportunity. Although the sensors seem to be reliable, additional verification with alternative CO₂ sensors is needed.

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