



Detection of CO₂ leaks from Carbon Capture and Storage (CCS) sites with combined atmospheric CO₂ and O₂ measurements

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Charlotte van Leeuwen and Harro A.J. Meijer

Centre for Isotope Research (CIO) Energy and Sustainability Research Institute Groningen (ESRIG) University of Groningen, the Netherlands





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 CO_2 is captured from (power) plants and stored underground in depleted oil and gas fields or deep saline aquifers.



Figure source: ZEP (Zero Emissions Platform)



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Situation in the Netherlands

Partially empty gas fields present, ideal storage for CO₂ Densely populated region, multitude of (fossil) CO₂ sources present

complicated detection of possible leaks



Edgar CO₂ emissions (2010)



Detecting CCS leaks in the atmosphere is difficult

- Rapid mixing of emitted CO₂ with the surroundings
- High natural variability of the atmospheric CO₂ concentration



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Possibilities for CO_2 leak detection: Use of tracers

- Add a tracer (e.g. SF₆, CH₄, perfluorocarbon)
 - Additional costs
 - Gases are strong greenhouse gases themselves
 - Migration of the tracers through the underground is not exactly the same as the migration of CO_2
- Use of natural tracers (¹⁴CO₂, ¹³CO₂)
 - ¹⁴CO₂ measurements are very expensive and only possible by flask measurements
 - ¹³CO₂ only works when:
 - significant difference between the $\delta^{\rm 13}{\rm C}$ of the biosphere and the source of ${\rm CO}_2$
 - sufficient CO₂ perturbation caused by the leak



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Measuring CO₂ at multiple locations



Two locations measure different signal. Yellow = background. Black = leak detection.

Main drawback: not possible to discriminate between a random (biospheric or fossil fuel combustion) point source of CO_2 and a leak of CO_2 .

This method was also applied on pipeline monitoring and published in: Van Leeuwen, C., Hensen, A. Meijer, H.A.J. (2013) Leak detection of CO_2 pipelines pipelines with simple atmospheric CO_2 sensors for carbon capture and storage. Int. j. greenhouse gas control 19, 420-431



Combined O₂ and CO₂ measurements

Most processes show an inverse relationship between O_2 and CO_2 , but CO_2 leaks have no counterpart in O_2



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O₂ and CO₂ together form the ideal leak detector Disadvantage: precise and accurate O₂ measurements are complicated

O₂ for leak detection: design criteria

- precision down to the ppm level
- long term calibration is less of an issue
- sturdy design, needs to be "mobile"
- fully automated, remote control
- should be able to run for about a month between services (air dryer and reference gas cylinder should last that long)



Design of a transportable $O_2 - CO_2$ instrument

Our system would fit in any small building or van.



the equipment in three "flight cases" (or road cases)



the small air inlet mast (6 m)





the reference and calibration cylinders with protection cap

-60° C drying system, with $Mg(ClO_4)_2$ trap follow-up

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The CO₂ / O₂ /(and δ^{13} C) device: under the hood



Quality of the measurements



	CO ₂ (ppm)		O ₂ / N ₂ (per meg)	
	Target 1	Target 2	Target 1	Target 2
Average stdev within a target run (n = 48)	0.011	0.010	12	15
Stdev of all target runs (n = 48)	0.021	0.018	8	8
Stdev of pairs, averaged (n = 47)	0.011	0.009	5	6



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Demonstrations in the field, at our station Lutjewad



station Lutjewad

CO₂ release experiments: 3-5 g/s



testing the wind



the small mast



CO₂ cylinder pack



short term day-time CO₂ release experiments



24-hour CO₂ release experiments



24-hour CO₂ release experiments





the three daytime releases: O_2/N_2 plotted against CO_2





CO₂ release tests: analysis

the two 24-hour releases: O_2/N_2 plotted against CO_2



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leak detection qualitatively very clear. Quantitative analysis?

I Compare time intervals (e.g. hours) with the general relationship



Strategies for analysis

- now calculate the average value and scatter around the slope for every individual hour of the whole period



- we can reformulate $(\mu_{hour} + \sigma_{hour}) \ge (\mu_{general} + \alpha \times \sigma_{general})$ into: $\alpha = \frac{(\mu_{hour} + \sigma_{hour})}{\sigma_{general}}$
- and then attribute an α value to each individual hour





Strategies for analysis



With this strategy, persistent leaks can be identified automatically detection level \approx 6 ppm (α =2 times observed varibility of 3 ppm)



II) Calculate the slope of time intervals (e.g. 6 hours) throughout time (without outlier filtering)



- Most of the time the slope will be around -5 per meg / ppm
- In case the slope is significantly higher (with a small error) a leak is identified



Two strategies for analysis



Three categories:

(1) slope fit error < 0.7 per meg/ppm, and slope > -1.5 per meg/ppm: leak
(2) slope fit error > 0.7 per meg/ppm and variability of CO₂>2 ppm: inconclusive

(3) all others: no leaks



Two strategies for analysis



Conclusions

- Combined O₂ and CO₂ measurements are a strong tool in detecting CO₂ leaks from a CCS site and the only tool to discriminate between a leak and another CO₂ source
- Our transportable system can be moved easily from one site to another
- The two analysis methods demonstrated are easy to automate, and together have a high potential for leak detection
- Their set points have been optimised for our system during our release tests; for a system with different precision these set points might need to be adapted
- The precision for O_2 can still be improved. The detection limit might be lowered then from ≈ 6 ppm to ≈ 3 ppm in these surroundings (with both many biogenic and anthropogenic sources of CO_2)
- The best strategy for leak monitoring is:
- Deploy a large number of cheap CO₂-only sensors (van Leeuwen et al., 2013. Ijggc. 19:420–431) (or alternatively one integrated large pathlength CO₂ sensor)
- When there is leak suspicion: bring in the CO₂-O₂ system



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