Chapter 6
Conclusions and outlook

This thesis presents two strategies to detect leaks of carbon dioxide (CO_2) from carbon capture and storage (CCS) sites in the atmosphere. Both strategies have their own advantages and drawbacks. The method that was presented in chapter 2, that uses multiple simple CO_2 sensors at close distance from each other, is relatively cheap compared to the much more sophisticated method that was presented in chapter 4, where atmospheric CO_2 measurements are combined with atmospheric oxygen (O_2) measurements. In a real CCS monitoring setup, a combination of the two strategies is probably the ideal solution.

As was discussed extensively in chapter 2, the relatively simple CO_2 sensors used here (Vaisala Carbocap GMP343) are in need of a laboratory calibration and temperature characterization every 3 – 6 months. As this is very labour-intensive and thus expensive, field data could also be used to determine the required corrections. This would be significantly easier and more accurate in case one or several high-quality CO_2 sensors are placed in the field as well. The transportable O_2 – CO_2 instrument presented in chapter 3 and 4 could provide this highly precise and accurate CO_2 data record and would be an excellent addition to a simple CO_2 sensor grid. (Significant) sources of CO_2 would be easily detected in the small-scale grid and the O_2 measurements could then discriminate random (fossil) sources of CO_2 from actual leaks. Instead of using multiple cheap CO_2 sensors such as the Vaisala Carbocap GMP343 one could also deploy a smaller amount of CO_2 sensors with a higher precision. Alternatively, the CO_2 concentration can also be measured as an average over a large area instead of at a single position by using long-distance open path laser spectroscopy techniques. Such instruments, using different techniques, have been demonstrated recently (Daghestani et al., 2015; Griffith et al., 2015; Schütze et al., 2013). Compared to an array of small, cheap CO_2 sensors as presented in this thesis, this approach has the advantage of requiring only one instrument, but the disadvantage of providing long-path averages.

The O_2 measurements can be performed at one or several fixed locations, but one could also deploy a transportable instrument such as the one presented in this thesis and bring it to suspicious locations if necessary. One single atmospheric leak detection strategy cannot be given: the strategy should be adapted to the specific area that needs to be monitored. Using atmospheric O_2 measurements for CCS leak detection does, however, always have the benefit that it can discriminate between random, fossil, sources of CO_2 and a real leak.

At the moment, atmospheric CO_2 leak detection is not very relevant in Europe, as all plans for onshore CCS were cancelled in the last years. The six large-scale CCS projects currently in operation or in under development in Europe all involve offshore storage of CO_2 (Global CCS Institute, 2014). In case CO_2 is leaking from an offshore storage location, the escaped CO_2 will largely dissolve in the seawater before it reaches the atmosphere. Leak detection in the atmosphere is therefore not possible for offshore CCS. The techniques presented in this thesis are, however, still interesting as they could also play a role in pipeline monitoring for pipelines transporting CO_2 partly over land towards an

offshore storage location and they can be interesting for CCS projects elsewhere in the world or maybe also in Europe in the future.

The core of this thesis is the development of the transportable O_2 – CO_2 instrument to aid in CO_2 leak detection at CCS sites and the demonstration of the technique by several CO_2 release experiments. The developed instrument is not only interesting for CO_2 leak detection, but also has wider merits for the atmospheric O_2 measurement community as a whole. The instrument is not state-of-the-art in terms of precision, but has several characteristics that could be useful in other applications. At the moment, the continuous O_2 – CO_2 instrument at the North Sea gas production platform F3 is partly broken due to contamination issues (mainly dust) of the instrumentation. The newly developed, transportable, and robust system will probably replace the old system at F3 in the near future. The protective cases will be highly advantageous at a remote (but sometimes crowded) location such as F3, as they protect all instrumentation against dust and accidental damage. Another advantage of replacing the system at F3 is the superior CO_2 sensor of the new system. Unlike at Lutjewad, there is no additional continuous CO_2 record at F3, so having a more precise data record is an advantage on this location. The air-drying system that is already present at F3, using two -80°C coolers should be used instead of the drying system of the transportable system. There is no need at F3 to compromise in the drying system and besides that, the existing system is also used to dry the air needed for flask sampling. The drying system should be able to operate without servicing as long as possible in this remote location. Using the original drying system (which also includes a Nafion column) will probably improve the precision of the measurements of the transportable system, as using a -80°C cooler seems to be better than a -60°C cooler combined with a magnesium perchlorate drying tube.

The transportable design of the new instrument could also aid in several other things such as studying the influence of wind direction and speed on the O_2 – N_2 fractionation at the air inlet. The transportable instrument could be easily brought to a wind tunnel to investigate the effects. The system can also be used for field campaigns, where (additional) measurements of atmospheric O_2 are needed.

Atmospheric O_2 measurements at the CIO are of sufficient quality, but unfortunately not state-of-the-art in comparison to some other laboratories. To get (back) to the group of top laboratories in the field, first of all the measurements of high-pressure gas cylinders need to be improved. This would especially improve the accuracy on the international scale, both for the flask sampling records and the continuous instruments. At the moment, air inlet procedures from the gas cylinders to the DI-IRMS are improved, reducing the influence of the regulators on the measurements. The tests were started after finishing the analysis of the data presented in this thesis and could therefore not be included. The first results, however, look promising, indicating a factor of two improvement in precision. Besides improving the gas cylinder measurements, more attention to the measurements in general is necessary. Big gaps in the data (e.g. the gap in the flask sampling record of Mace Head around 2009 and the gap in the continuous data record at Lutjewad in 2013 – 2014) form serious problems for trend and seasonal cycle fitting. Gaps (of all sizes) should be prevented as much as possible. Availability of experienced technical and scientific staff is crucial to achieve this, but is unfortunately often lacking due to money shortages.

Continuous atmospheric O_2 measurements appear to be even more challenging than flask sample measurements. The equipment is extensive and fragile (although less so for the design in this thesis) which can lead to broken parts (e.g. at F3) and the measurements are easily influenced by temperature fluctuations in the surroundings of the instrument (e.g. at Lutjewad for both the fixed as well as the transportable instrument). The extensive gas-handling scheme is also prone to leaks and they can be hard to discover. To improve the continuous atmospheric O_2 measurements at the CIO, several recommendations can be given. First, the temperature of the surroundings of the instrument needs to be as stable as possible: both short-term and long-term fluctuations need to be minimized. Secondly, careful attention should be paid to the high-pressure gas cylinders. They need to be replaced well on time to avoid drift of the measurements when they run nearly empty. The regulators also need careful attention, as they seem to influence the measurements of the continuous instruments, just as they influence measurements on the DI-IRMS. Aspirated air inlets (Blaine et al., 2006) should be installed to prevent thermal fractionation issues that could influence the O_2 measurements (Manning, 2001). To be able to identify potential problems in the sampling procedure at an early stage, two sample lines must be installed instead of one, as is done by e.g. (Manning, 2001; Wilson, 2012). The two (identical) lines should be switched on a regular basis and if everything functions well, no difference should be visible between the two lines. An increase of the data output frequency would also be a big improvement of the system. To do this, the switching time should be reduced as much as possible. For the transportable system the switching time was already reduced from 5 to 3 minutes but this could even go down to one minute as is the case in Weybourne atmospheric monitoring station (Wilson, 2012). This would require an adaptation of the CO_2 sensor from one to two paths, such that the reference air has its own path and switching between the two lines is not necessary anymore. In such a setup, switching between sample and reference air is only necessary for the O_2 measurements.

The flask sample records of Lutjewad and Mace Head now both cover a time period of about 15 years. Careful attention is, however, also needed for the flask sampling records, as contamination issues should be discovered as soon as possible to prevent big gaps such as the one in 2009 at Mace Head. Both flask-sampling programs at Lutjewad and Mace Head will be continued in the future. In addition, a new atmospheric O_2 flask-sampling program has been started at Halley station, Antarctica. As this station is extremely remote, flasks are sent (and received back) only once a year. Preventing leaks during sampling, and especially during long-term storage of the flasks, is therefore even more important here than it is at other locations. The flasks from the first year of the campaign just arrived back at the laboratory of the CIO in Groningen and first results are expected soon.

6.1. References

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