Summary

Methods for observation and quantification of trace gas emissions from diffuse sources

This thesis is a synthesis of experimental studies that aims to contribute to the improvement of estimates of trace gas emissions from diffuse sources into the atmosphere. The term diffuse source is used to distinguish from sources that do have well defined exhausts like for example chimneys or ventilators. Diffuse sources include:

- soils, either natural, agricultural or industrial (landfills);
- groups of sources within a confined area (farm with multiple animal houses, manure storages or industrial plants with a large number of point sources;
- regions: a group of farms; farms and industries, urban areas.

Work is presented for the main greenhouse gasses Carbon dioxide (CO_2), Methane (CH_4) and Nitrous oxide (N_2O). Ammonia (NH_3) is subject for research, as an important air pollutant that is mainly emitted from agricultural diffuse sources.

About 20% of the total GHG emission to the atmosphere is emitted from diffuse sources either as CO_2, CH_4 or N_2O. The uncertainty in the emission level from these sources is a dominant component in the uncertainty of the national emission level for the Netherlands. For ammonia 88% of all emissions in the Netherlands originate diffuse source systems, mainly in the agronomic sector. In the 27 EU countries the rate is even higher, 93%. Similar to the greenhouse gas case, the NH_3 emission level on a national and European scale is still far from certain.

The main reason for the large uncertainties in the emissions from diffuse sources is the difficulty to quantitatively measure these emissions that can show large variations both in time and in space. Depending on the trace gas species different sensors and different measurement methods are required to evaluate the emission levels. This thesis gives an overview of the sources, the emission levels and the associated uncertainties. Also currently available measurement sensors and methods are described.

Examples of studies that aimed to develop measurement strategies for diffuse source systems are provided in the five publications that are part of this thesis. This set starts with two papers on the micrometeorological technique known as the relaxed eddy accumulation technique (REA) which was evaluated for carbon dioxide (CO_2) and for ammonia (NH_3). This technique measures the net vertical gas transport above the surface. The emission from a diffuse source system can also be evaluated remotely by measurement of gas that is transported away from the source area with the wind in a gas plume. Two papers discuss the use of this type of measurements for evaluation of methane emissions from landfills and for the assessment of NH_3 emissions from a farm site in Germany. The last paper describes how the plume technique with mobile measurements was applied to quantify methane emissions from farms. After these examples a synthesis is made both from the technical perspective and from the perspective of different source types.

General remarks
Diffuse source systems have challenged scientist for several decades. Although our knowledge has improved considerably, science has still far from unravelled how diffuse source systems work at the process level and how to quantitatively measure the net emission on an annual basis on representative spatial scales (for example > 1ha ecosystem, full farm or full landfill). This holds for example for the net CO_2 equivalent exchange after land-use change, for the direct and indirect N_2O emissions from fields and

In this thesis three improvements to better understand emissions from diffuse source systems were explicitly demonstrated.

- The fast box measurement that makes a combination of a simple box method with an advanced concentration sensor (with high resolution both in concentration and in time). This method helps to tackle spatial variability of the emissions within a given source area. It also helps to show the non linearity of the concentration increase in box measurements discussed above.
- The plume measurement that was used to provide source integrated emission data both for individual farms and for landfills.
- The relaxed eddy accumulation for flux measurements offers a temporal solution for components were fast eddy covariance measurements are still not possible.

Conclusions from a source perspective
Important diffuse source systems that feature in this thesis are landfills, agricultural fields and farms. For landfills, the data obtained with plume measurements can be considered important internationally. Thus far emission accounting methods for these sources are not standardized. Within landfill research there is a hypothesis that the use of different methods leads to a factor four difference between the methane emission estimates for an average UK landfill compared with a similar Dutch landfill.

For agricultural fields, there is still a large uncertainty both in direct and indirect emissions of N_2O. For these source systems both the fast box method and new eddy covariance data should provide better emission estimates. To evaluate the net greenhouse gas balance, there is an evident need to perform simultaneous and co-located measurements for CO_2, CH_4 and N_2O. This is now possible. It is remarkable that in the Netherlands there is virtually no data with co-located NH_3 and greenhouse gas measurements. This is strange because our country has the highest emission densities for both greenhouse gases and NH_3 in Europe.

As for farms the use of farm integrated emission measurements are still hardly used. The plume measurement data obtained thus far did however contribute to the debate on the CH_4 emission factor for dairy cows. The result was a significant update of the emission calculation method for this source.

There is no such thing as “the best method” to perform the emission evaluations for any of these source types. There is even a potential danger of systematic biases when using a single method and rely completely on more and more measurements of the same type to reduce the uncertainty. On the other hand, comparability of different experiments is an advantage of using the “standard” methods. The best option at the moment seems the use of a combination of techniques in order to better quantify the emissions from diffuse source systems. Emphasis should be on further development of the more integrating (micrometeorological) techniques to enable a significant reduction in the uncertainty to whatever is agreed on as an acceptable level.

Finally
This thesis shows the slow but steady progress over almost two decennia in this field of science. In the decades to come the key to further improvement is in new, improved or significantly cheaper sensors that will enable further improvement available strategies or even new measurement strategies. As for the national and global scale combination of ground truth measurements and satellite retrievals will be a crucial step. For individual sources, further development of the plume methodology towards fence line monitoring systems is a likely development. The latter implies further development of low cost sensors, lidar (light detection and ranging) systems or other remote techniques. The fundamental errors for box and mass balance method of course have to be resolved as

water bodies, for methane emissions from landfills and both for methane and NH_3 emissions from livestock activities.
The uncertainty of net gas exchange between diffuse sources and the atmosphere is partially explained by the large number of individual source systems in combination with the fact that different geographical position, environment, weather and management can have a strong impact on the emission processes. The multitude of source locations makes it inevitable to extrapolate emission measurements. Results obtained at a limited set of locations have to be used, to calculate the emission over the full extent of the source type. This will require a combination of data and parameterizations in computer models. Meteorological data, land use and management data statistics as well as distributions in space and time of emission driving parameters are needed to do this extrapolation.

The parameterizations used in the computer models have to be generated (or validated) with experimental work. Zooming in on the emission measurements, there are two parts, the gas concentration measurement itself and the method used to translate these measurements into a flux. These two parts are interlinked. Some methods, like for example micrometeorological techniques, can only be carried out with fast response sensors that can show small changes in ambient concentration levels. The slow but continuous improvement of sensors makes currently used methods more accurate. Apart from that also enables the development of innovative measurement strategies.

Conclusions from a technical perspective
With the current state of sensor techniques, the conclusion is that fundamental methodological issues for several measurement methods used are still not resolved. Two examples are discussed:

- Mass balance measurements that are both underlying the national emission estimates for CH_4 from landfills and NH_3 from fields treated with manure have provided overestimates (probably between 5-15%) in the results thus far. The reason is that the available sensors used for this method cannot resolve the so called backward turbulence correction needed for this method. When implemented, this correction will always provide reduced flux levels.
- A similar reasoning holds for the box measurements that in general provide underestimates of the real flux by between 0-30%. An explanation for this is that the available sensors are too slow. Most experiments are carried out at a time resolution that prevents the detection of non-linearity in the concentration increase under a box. Consequently the bulk of the box measurements reported in literature used linear interpolation to calculate the emission level. And these estimates are very likely to be underestimations of the real flux.

These two “errors” have a very similar background. The need for correction in both cases was already discussed 20 years ago. With available sensors however the correction can hardly be carried out. So almost no one uses it. This would not be much of a problem when these errors were random. But unfortunately the corrections needed lead to a systematic shift in the results. New sensors can now show and quantify these errors. There are several recent publications discussing the box measurements. For the mass balance method a similar discussion will have to start.

soon as possible and if possible correction algorithms for historic data have to be developed.

This thesis wants to emphasise the importance or measurements and innovation of measurement techniques. Measurements alone do not provide all the answers. Activity data, modelling tools and measurements from a trinity needed to evaluate diffuse source emission levels. Direct observations, however, are leading within this trinity. Improvement of sensors and measurement techniques are therefore important to better quantify the emissions of trace gasses from diffuse source systems.

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