Cabauw greenhouse gas measurement 2015

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Acknowledgement

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Contents

Summary

At the Paris climate meeting in December 2015, 196 countries agreed to limit greenhouse gas emissions in order to keep the world wide temperature change increase within 2 degrees. To reach this goal, the aim is to have net zero emissions for CO_2 equivalent gasses in 2050. This aim, will lead to extra pressure to reduce fossil fuel related emissions and a quest for options to sequester or store extra CO_2 from the atmosphere. The need to understand where the greenhouse gasses come from and where they end up thus is more prominent than ever. This is why greenhouse gas concentration measurements in the atmosphere, that show both the effect of emissions and the potential response of natural systems to climate change, are so important. Moreover, taking into account the timescale on which climate change occurs, there is the need for long time series that document the gas exchange between oceanic and terrestrial sources, sinks and the atmosphere. This report shows the unique data trend obtained at the Cabauw tall tower that provides measurements over the last 22 years. In 2015, a serious backlog of data processing was solved with aid of the INGOS EU program. The Cabauw dataset is made available to the international community through the Obspack NOAA project. Within the national project for 2015 a new data interpretation tool, the Cabauw emission indicator is proposed. From the first results the following preliminary conclusions can be drawn:

- The Cabauw indicator confirms the trend in the Dutch CO_2 emission (which is assumed to have only a 3% uncertainty in the emission registration estimates).
- The Cabauw indicator does not support the emission reduction for CH_4 and suggests that there is hardly any decrease. Which is in line with the results of the inverse modelling calculations that indicate the Benelux emission level for methane might be 40% above the estimated level.
- The indicator calculations corrected for the boundary-layer height revealed that as a result of an increase in boundary-layer height the concentration levels have increased less prominent over the last 20 years which is important for evaluation of air pollution data.

More research is needed to improve this evaluation tool in the coming year. Furthermore, in 2015 the Cabauw hardware was changed substantially with the installation of a new fast Fourier transform infrared spectrometer. This new instrument will further improve the quality of the data obtained and facilitate better assessment of sources and sinks calculations for the Dutch territory and beyond.

Samenvatting

Op de Parijse klimaattop in december 2015 besloten 196 landen om de uitstoot van broeikasgassen te beperken om zodoende de wereldwijde temperatuur verandering binnen 2 graden te houden. Om dit doel te bereiken is een netto-nul-uitstoot van CO_2-equivalent gassen in 2050 nodig. Dit zal leiden tot extra druk op fossiele brandstofuitstoot en een zoektocht naar mogelijkheden om CO_2 af te vangen of op te slaan om extra CO_2 uit de atmosfeer te onttrekken. Met dat doel is de noodzaak om te begrijpen waar de broeikasgassen vandaan komen en waar ze uiteindelijk terecht komen prominenter dan ooit. Dit is de reden waarom de metingen in de atmosfeer, dat zowel de emissies als de mogelijke reactie van natuurlijke systemen op de klimaatverandering toont, zo belangrijk zijn. Bovendien, gezien de tijdschaal waarop klimaatverandering optreedt, is er een noodzaak voor lange tijdreeksen die gasuitwisseling tussen oceanische en terrestrische bronnen, putten en de atmosfeer documenteren. Dit rapport toont de unieke meetserie, die de trend in broeikasgassen boven Nederland documenteert, over de afgelopen 22 jaar. De metingen werden uitgevoerd op de 200 meter hoge Cabauw toren. In 2015 is de achterstand met gegevensverwerking opgelost met behulp van het INGOS EU-programma. De Cabauw dataset is nu internationaal beschikbaar door het Obspack NOAA project. Binnen het nationaal project voor 2015 is, als een nieuwe interpretatie van gegevens, de Cabauw emissie-indicator ontwikkeld. Uit de eerste resultaten kunnen de volgende voorlopige conclusies worden getrokken:

- De Cabauw indicator bevestigt de trend die wordt gerapporteerd in de Nederlandse CO_2 emissieregistratie (waarvan een onzekerheid van slechts 3 % wordt verondersteld).
- Voor CH_4 bevestigt de Cabauw indicator niet de dalende trend die zichtbaar is in de emissieregistratie, met een indicator die nauwelijks daalt. Dit resultaat is in lijn met die van inverse modellen die laten zien dat de emissies van CH_4 in de Benelux mogelijk 40 % boven de veronderstelde emissies liggen.
- De resultaten voor een grenslaaghoogte gecorrigeerde indicator laat zien dat door een toename in de grenslaaghoogte de concentraties van broeikasgassen minder prominent zijn gestegen in de afgelopen 20 jaar. Dit resultaat is belangrijk bij de analyse van luchtvervuilingsdata.

Meer onderzoek is nodig om de methoden waar mogelijk te verbeteren in 2016. Hiernaast is in 2015 is de hardware op Cabauw aanzienlijk veranderd met de installatie

van een nieuwe snelle Fourier-transformatie infrarood spectrometer. Dit nieuwe instrument zorgt voor een verdere verbetering van de kwaliteit van de verkregen gegevens en vergemakkelijkt een betere beoordeling van de bron en put berekeningen voor het Nederlandse grondgebied en daarbuiten.

1

Introduction

At the climate top in Paris (December 2015) organized by the United Nations Framework Convention of Climate Change, 196 countries agreed that the average global temperature cannot rise more than 2^{o}C. Making this the first ever global agreement to reduce climate change. In order to do so the countries have agreed to decrease emissions of greenhouse gases (i.e., carbon dioxide and methane). In fact, the countries agreed to limit the amount of anthropogenic emissions by 2050 to levels that can be absorbed by natural sinks. There is however no implementation plan how every country will achieve that. It is not unlikely that in the Netherlands also a further reduction of greenhouse gas emissions will be implemented.

In order to reach the ambitious goals, measurements of greenhouse gases are crucial. Measurements keep track of greenhouse gas concentration levels and they can also be used to independently estimate the emissions of greenhouse gases. Furthermore, measurements are essential to determine the size of natural sinks.

At Cabauw (located in the centre of the Netherlands), the first greenhouse gas (GHG) measurements on the tall tower (200 m) started in 1992 and 1993 with carbon dioxide (CO_2) and methane (CH_4). Later on, measurements of nitrous oxide (N_2O), carbon monoxide (CO), and sulphur hexafluoride (SF_6) were added. Radon measurements that started in 2005 allow an assessment of the difference between the exchange of greenhouse gas over sea and land. The ECN GHG data, together with the full suite of meteorological parameters as measured by the Royal Netherlands Meteorological Institute (KNMI) and air pollution data from the Dutch National Institute for Public and Health (RIVM) provide a unique data set that shows how Dutch greenhouse gas concentrations are changing over more than two decades. Thus, this data provides an essential factual foundation to support the climate change debate. The measured concentration levels have inherit information about the emission levels of greenhouse gas sources within the Netherlands and North-western Europe.

2

Methods

2.1 Site description

The measurement site is located in the centre of the Netherlands, near the village Cabauw. The geographical position is 51°58'16' N, 4°55'36' E. (see Figure 1~~Figure 1~~). The surrounding landscape is composed of meadows and ditches, with scattered villages and a flat surface within a radius of at least 20 km. About 1 km south of the tower runs the river Lek (part of the river Rhine system). The subsurface consists of a river clay (0.4 to 0.8 m) on top of a peat layer. The water table at the Cabauw site is usually 1 m below the surface, but can be considerably higher during wet periods. The Cabauw site is owned by The Royal Netherlands Meteorological Institute (KNMI) and boundary layer meteorological observations have been carried out at the site since 1972. The main tower is 213 m high. A detailed description of the facilities is given in Vermeulen et al. (2009).

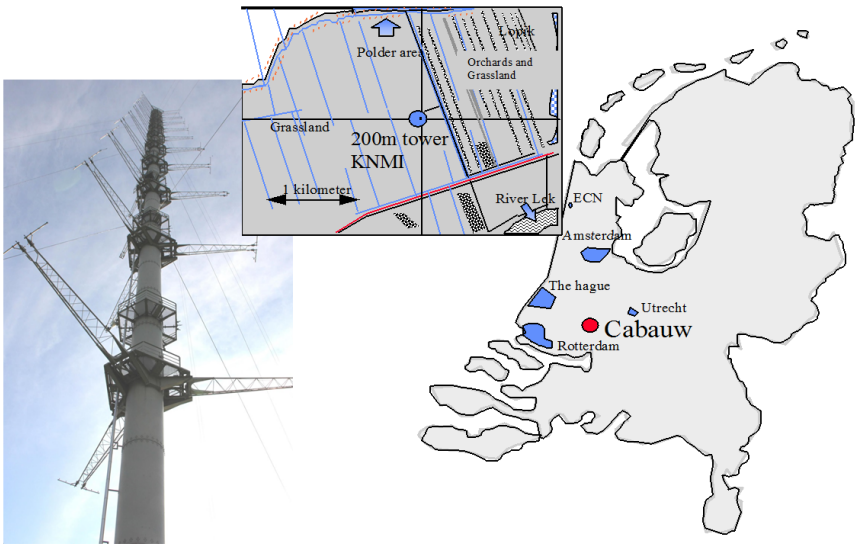

Figure 1: Cabauw tower location and photograph. The total height of the tower is 213 m.

2.2 Measurement setup

The greenhouse gas concentration measurements of ECN take place on a 213 m high meteorological tower near the village of Cabauw. The analytical equipment for the greenhouse gas measurements is placed in the basement of the tower. Ambient air is sampled continuously from 200 m, 120 m, 60 m and 20 m height to the basement. Meteorological data are provided by the KNMI. With steps of 20 m wind speed, wind direction, pressure, temperature, radiation and relative humidity are available.

Figure 2: Instrumental setup for the CO_2 measurements at the Cabauw cellar in 1995. On the left calibration cylinders. In the middle the Siemens Ultramat NDIR, the data logger, and switching valves for height selection. On the right the central processing unit with connection to ECN.

The Cabauw data set can be divided into four distinct periods, with measurements of different gases, at different heights, and with different instruments (Period A; 1992-1997, Period B; 2000-2004, Period C; 2004-2015 and Period D; 2015-now). The years 1997-2000 are lacking due to a major overhaul of the Cabauw tower. The first three periods (A till C) are described in detail by Vermeulen et al. (2011) .

Measurement of CO_2 and CH_4 at 200 m height were initiated in 1992. In order to evaluate the CO_2 exchange over the field surrounding the Cabauw tower CO_2 was also measured at 1, 2, and 10 m height (see Hensen et al., 1995, Vermeulen et al., 1997, and Hensen et al. 1997). A Siemens Ultramat Non Dispersive Infra-Red monitor (NDIR) was installed for CO_2. A Gas Chromatograph (GC) system (Carlo Erba GC8000) equipped with a flame ionization detector (FID) was installed for CH_4. The sampling air was drawn through Teflon tubes (PTFE) with a flow of 30 L min^{-1}.

Period B starts in 2000, after a major overhaul of the Cabauw tower. CO_2 and CH_4 data is from then onwards measured at four heights (20, 60, 120, and 200 m). Sampling air was drawn though polythene tubes with a flow of 20 L min^{-1} and a nafion prY-dryer was installed after the inlet.

During period C, measurements of, N_2O and SF_6 measurements were added in 2004 and in 2007 CO and H_2 were added. The Siemens NDIR was replaced by a Licor 7000 NDIR for CO2 and a new GC system (Agilent 6890N) equipped with an FID for CH_4 and an electron capture detector (ECD) for N_2O and SF_6. In 2007 an additional reduction gas analyser (RGA) was installed for both CO and H_2 (Popa et al. 2011). From 2011 onwards, the CO_2 and CH_4 data were also collected by a cavity ringdown optical measurement system (Picarro 2301) in parallel with and as a backup of the NDIR (CO_2) and GC (CH_4). Due to instrumental issues continuous records of N_2O, SF_6, CO, and H_2 were difficult to maintain from 2013 onwards. A Fourier transform infrared spectroscopy analyser (Spectronus FTIR), able to measure CO_2, CH_4, CO, and N_2O was prepared in 2014-2014 and installed in January 2015. This FTIR replaced the GC system as well as the NDIR. Sampling air was drawn though Synflex 1300 tubes with a flow of 10 L min^{-1} and a nafion pry-dryer was installed after the inlet.

A new period (D) starts in August 2015 where both the instrumental setup as well as the sampling of the air is changed. Both the new Spectronus and the Picarro sample from 20L buffer volumes instead of 'point' sampling (direct withdrawal of air from the sampling lines). Sample air is, like in period C, drawn through Synflex 1300 lines with a flow of 10 min^{-1}. The buffer volumes represents an average of the inlet air of the last 20-30 minutes and improve the temporal data cover of the profile measurements. Before the buffer volumes, the profile measurement represent the inlet air for just a fraction of the averaging time (in general 30 minutes) as each inlet height is connected to the instruments for 5 minutes only.

From the time series of the greenhouse gas concentrations, emission levels can be estimated using different methods. In the following sections we first show two methods that were developed at ECN to estimate emissions: the HUMP method and the Cabauw emission indicator. The results of the Cabauw emission indicator will be compared to the bottom-up estimates of the emission registration carried out by the National Institute for Public Health and the Environment (RIVM).

2.3 The HUMP method

The HUMP method relies on the decoupling of different gas layers at different altitudes in the atmosphere during the night. This decoupling occurs because there is no longer energy input from the sun. Therefore, the Earth's surface starts to cool, which also results in a cool layer of air near the surface, making the atmosphere stable. The lowest layer of the atmosphere in which the exchange occurs between the Earth's surface and the atmosphere (i.e., boundary layer), becomes small (in the order of 100 m). With emissions continuing during the night, gas is trapped and accumulates close to the surface. For nights where the boundary-layer height is below the highest measurement at the 213 m tower a clear increase in the concentration levels is observed at the lower measurement heights. At the same time the concentrations at higher measurements heights stay more or less constant. In the morning, when the sun rises again the boundary-layer height will grow, the lower layer with enhanced concentrations will be mixed with layers above (i.e., there is no longer a decoupling between these layers in the atmosphere).

As the mixed layer grows the measurements at the different heights tend to decrease together. When the mixed lower layer reaches the next measurement level (e.g. 200 m level) an upward jump in the concentration at that height is observed. At that moment the height (H) of the well mixed box that contains the input due to emissions in the region over night is exactly 200m. The emission (E) can be calculated from the 200 m concentrations during the decoupling (C_{night}), the 200 m concentration after mixing occurs (C_{mixing}), and the time that the different layers are decoupled (t). The formula as described by Hensen et al. (2000) reads:

$$E = \frac{C_{mixing} - C_{night}}{H * t}. \quad (1)$$

The values of C_{mixing}, C_{night}, and t from the time series are derived from an algorithm (see Figure 3~~Figure 3~~). This algorithm still needs improvements in order to also identify periods where the 200 m concentration level is less stable than is the case for this example. At the moment without these improvements no results can be shown.

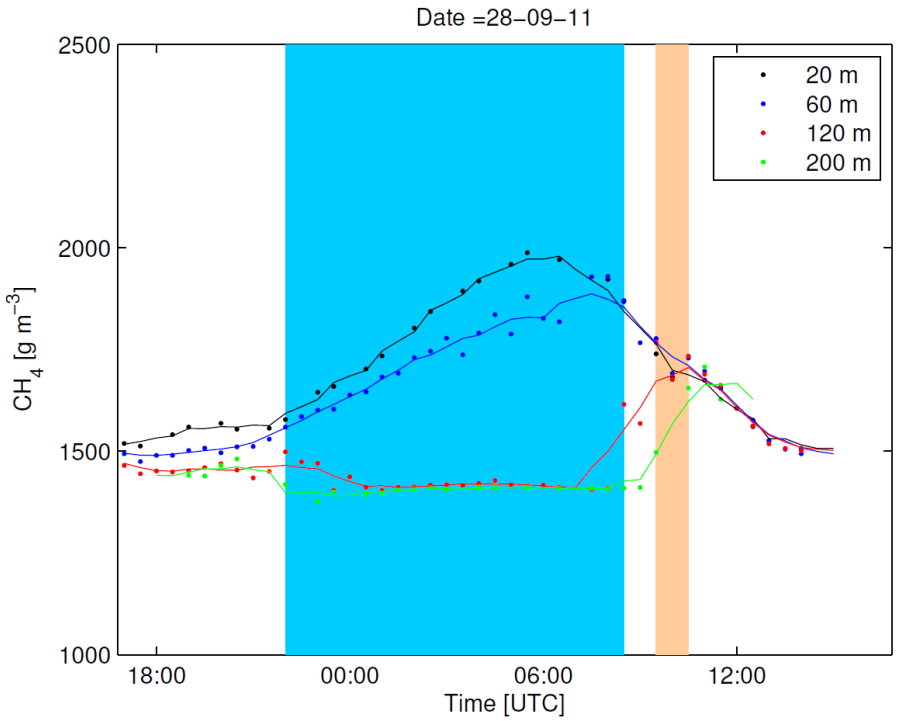

Figure 3: Decoupling during the night of the concentration levels of CH_4 at the four heights. Blue indicates the time period where the 200 m concentration is lowest and more or less constant. Orange indicates the time period where the 200 m CH_4 concentration rises rapidly in the morning to reach similar levels at the other measurement heights.

The main advantage of the HUMP method is that it quantifies the emission for a specific night. However, in order to do so the decoupling of the atmosphere has to occur and the boundary-layer height should be below the highest measurement level (200 m at Cabauw). This method is therefore more suitable to investigate specific events in more detail, but less efficient to estimate the average emissions level per month. In combination with footprint maps the method can give insight into the source area that is linked to the calculated emission level.

2.4 Cabauw Emission indicator

The Cabauw indicator is a simplified method to estimate the emissions from the 10th and 90th percentile of a month of measurements (see **Figure 4**~~**Figure 4**~~). The method has the potential to be applied in near real-time, with a time-lag of only two weeks. As the name already suggests, this method gives an indication of the emission only. It is not a qualitative value of the emission. The method, however, can provide information in terms of trends in time and thus show if emissions are going down or up.
The Cabauw indicator is calcaulte by subtracting the 10th percentile from the 90th percentile for every month. The 10th percentile is assumed to represent the 'regional background concentration'. The 90th percentile covers all data except the highest 10% of the data. It is assumed to include all contributions to the gas concentration on a regional scale on top of the 'regional background concentration'. The very local peaks

will be filtered out since they will fall in the top 10% of the concentration distribution over that month.

Greenhouse gas concentrations are sensitive to the boundary-layer height, especially at night (see spikes in Figure 4~~Figure 4~~). Therefore, any trend in the emission indicator can be influenced by the boundary-layer height. In order to investigate this influence the emission indicator is multiplied with the boundary-layer height. Unfortunately, boundary-layer height measurements are not available for the entire GHG time series. Therefore, the ERA-interim data (from the ECMWF model) of the boundary-layer height is used.

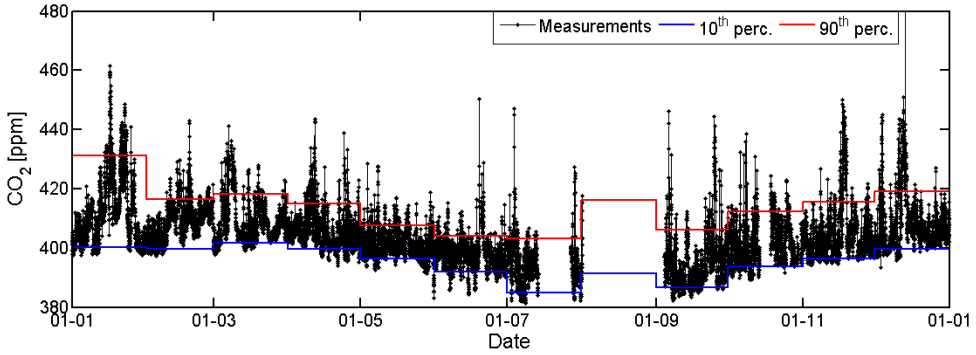

Figure 4: Time series of the 200-m level CO_2 with the monthly 10th percentile and the 90th percentile indicated for 2013.

2.5 Emission registration data

In order to evaluate the performance of the Cabauw indicator method the estimated emissions were compared with the total annual emission sover the Netherland which are available from emission registration by RIVM.

The emission registration as used by the RIVM has a certain amount of uncertainty. For example, the CO_2 emission caused by agriculture is relatively unknown. However, the emissions from agriculture are relatively low compared to other sources. Thus, RIVM estimates that the uncertainty in CO_2 emissions is 3%. Table 1~~Table 1~~ shows the estimated uncertainties. It is clear that both for methane and nitrous oxide, with estimated uncertainty levels of 25 and 50% respectively, independent emission estimates based on atmospheric measurements are useful.

Table 1: Uncertainty estimated by RIVM for the emission registration data (source: http://www.emissieregistratie.nl/erpubliek/content/explanation.nl.aspx).

Gas	**Uncertainty [%]**
CO_2	3
CH_4	25
N_2O	50
NH_3	17
NO_x	15

3 Results

3.1 Measurements

3.1.1 Concentration time series

An overview of the complete dataset collected so far is given in Figure 5~~Figure 5~~. The time series plotted show effects on different time scales, which is displayed in more detail in Figure 6~~Figure 6~~ for CO_2. On the decadal time scale the continuous increase of in this case CO_2 concentration is shown. This upward trend emerges on every measurement station around the world. The increase in concentration can be used to calculate what has changed in the radiation balance of the earth and how much infrared light emitted from the earth surface and atmosphere is being intercepted by greenhouse gasses.

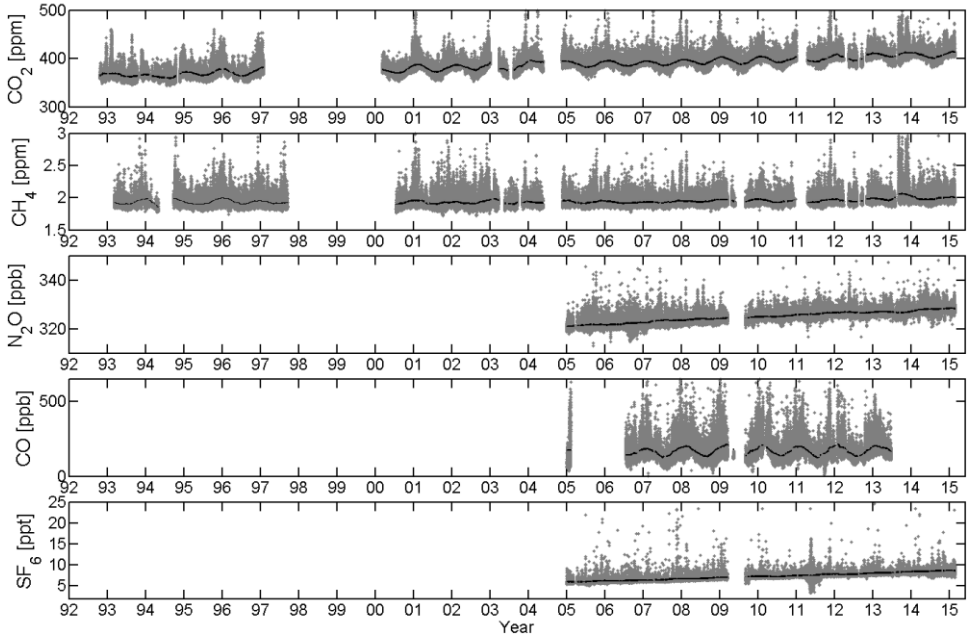

Figure 5: Overview of the greenhouse gas data set collected at Cabauw for the 200 m level.

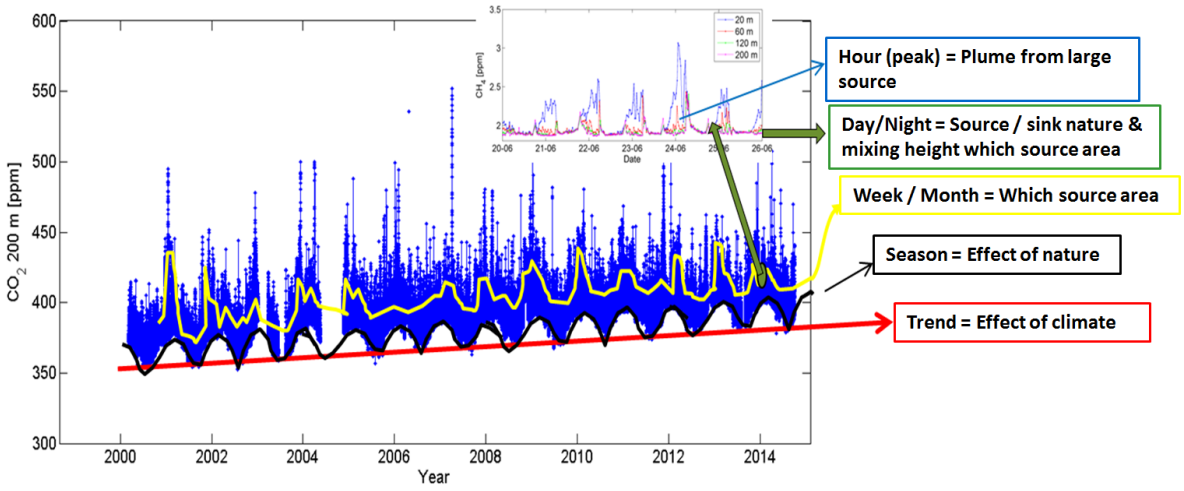

Figure 6: Time series of the 200 m CO_2 Cabauw data, with an indication of the contribution of different time scales to the series.

The sine-like movement overlying the rising line of CO_2 shows the seasonal variation in the concentration level. There are two processes generating this pattern. First, in summer the vegetation is a net sink of CO_2 as plants or plankton grow and take up CO_2. In autumn and winter a big part of this accumulated carbon is emitted back again into the atmosphere. Second, there is the seasonal amplitude in the boundary-layer height, which is the lower layer of the atmosphere where exchange between the earth's surface and the atmosphere occurs. This height makes a 'box' in which the emitted gas accumulates (see Chapter 2.3). In summer, this reasonably well-mixed layer in the atmosphere is between a few hundred meters and a few kilometres high, but in winter that height is in general smaller. With the same amount of gas emitted into the atmosphere, the measured concentrations are higher in winter than those in summer.

Concentration variations with a time scale in the order of weeks occur due to high and low pressure systems that determine the wind sector over Europe that has interacted with the air mass. Air that travelled over important source areas on the European continent accumulates more greenhouse gasses compared to air that travelled over the relatively clean North Sea.

On the diurnal scale a zoom of a few days is shown with individual measurements every 30 minutes for the four heights. The plot shows the effect of day-night variation, which is a very dominant process that can cause what seems to be 'noise' in the big picture. The high peaks on top of the lower base line show how during night the GHG concentration levels increase. In the absence of the solar irradiance the boundary-layer height (i.e., the lid on the box) decreases at night often somewhere between 50 and 300 meters. On top of the day-night pattern relatively short extra spikes for minutes or hours are caused by more local sources or larger event emissions.

In summary there is a lot of different information in the measured concentrations. The challenge is to retrieve this 'hidden' information on emissions from the data sets.

Although the greenhouse gas data set at Cabauw is extensive, for some components (e.g., CO and NO_x) the data period over which the data is collected is too short to do trend analyses (e.g., emission trends). It is important to realize that both emission reduction measures and climatic change occur on a timescale of decades. In order to document and show the effect or impact of these processes, long term continuous operation of greenhouse gas measurement sites is crucial.

3.1.2 Wind Rose

One way of getting a view of the emissions around Cabauw is from wind rose plots. A wind rose shows which concentration levels are measured from a certain gas for different wind directions. In principal, the concentration from a certain wind direction should be high when the emission from this direction is high.

The excess CO_2 concentrations, defined as the actual CO_2 concentrations minur the regional background (the yearly 10^{th} percentile) is shown as a wind rose for 1996 in Figure 7~~Figure 7~~ and for 2014 in Figure 8~~Figure 8~~. This visualisation makes sure different years can be easily compared to one another. For 1996, the excess concentrations from southwest are clearly higher compared to other wind directions. This reflects the longer path length that the air parcels cover over land. In the southeast the emissions from for example the Ruhr area in Germany will be visible. Low concentration of CO_2 is observed when the wind is blowing from the northwest, when relatively clean North Sea air is sampled.

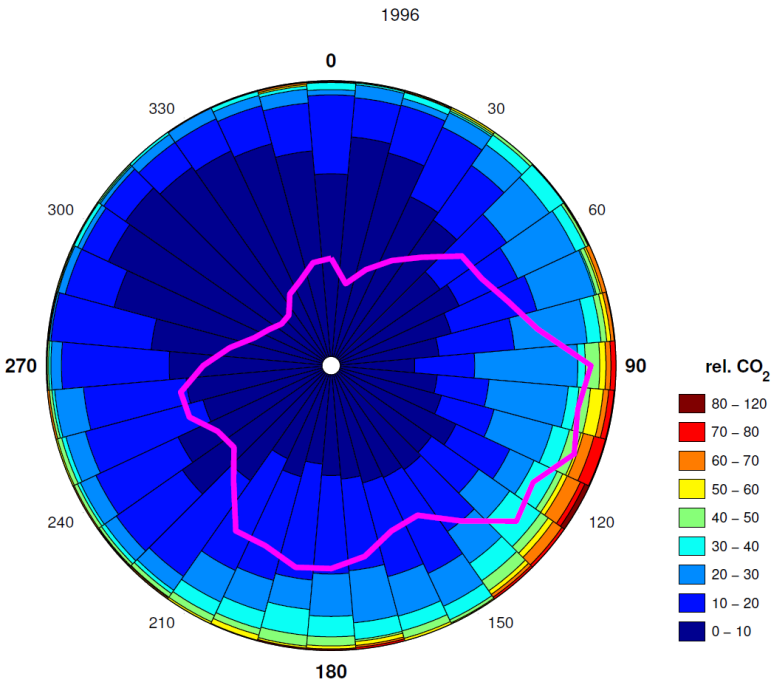

Figure 7: Wind rose of the CO_2 excess concentrations measured at Cabauw at 200 m in 1996. Shown in colour is a histogram of the excess CO_2 concentration (concentration – yearly 10^{th} percentile), normalized per wind direction. In pink on top is the average excess CO_2 concentration per wind section.

The same wind rose is plotted for 2014 in **Figure** 8~~**Figure** 8~~. The general pattern of the wind rose is the same with high excess concentrations in the southeast and northwest. However, especially the high peaks seem to have decreased in 2014 compared to 1996. Suggesting a decreased emission level in that sector, assuming that for example the boundary-layer height was similar in both years.

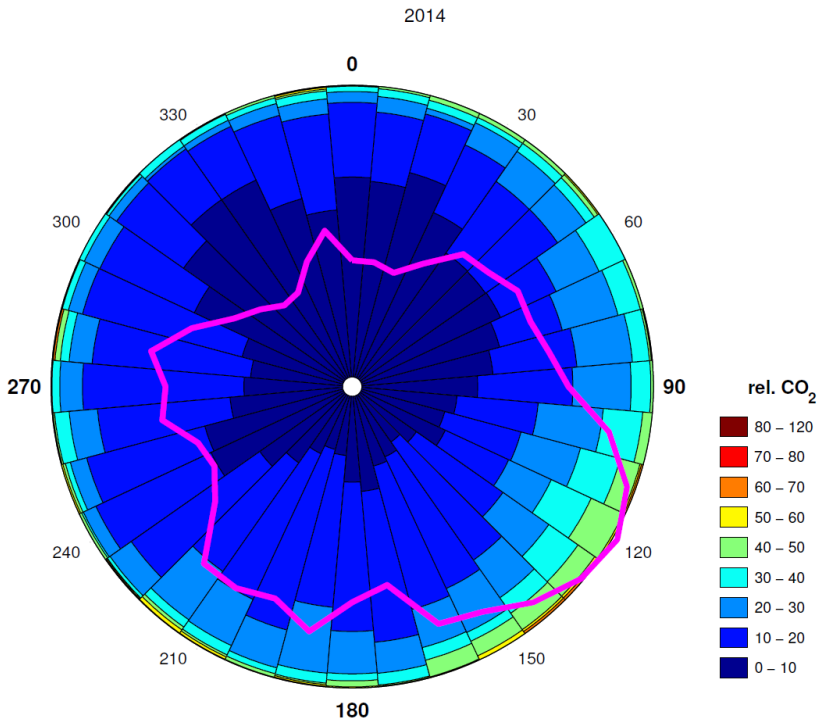

Figure 8: Wind rose of the excess CO_2 concentrations measured at Cabauw at 200 m in 2014. Shown in colour is a histogram of the excess CO_2 concentration (concentration – yearly 10^{th} percentile), normalized per wind direction. In pink on top is the average excess CO_2 concentration per wind section.

When both relative CO_2 concentrations are plotted on the same wind rose, as shown in **Figure** 9~~**Figure** 9~~, it is apparent that the mean excess concentrations are more or less the same in 1996 and 2014 (keep in mind that the absolute concentration levels have increased on the whole northern hemisphere) for the different wind directions. So, although the very high peaks in the southeast might have decreased, the mean excess CO_2 concentrations per wind directions are similar for 1996 and 2014. Apparently, there is a counterbalanced of more frequent peaks with a moderate CO_2 concentration (in the order of 30-40 ppm above the 10^{th} percentile) for the year 2014.

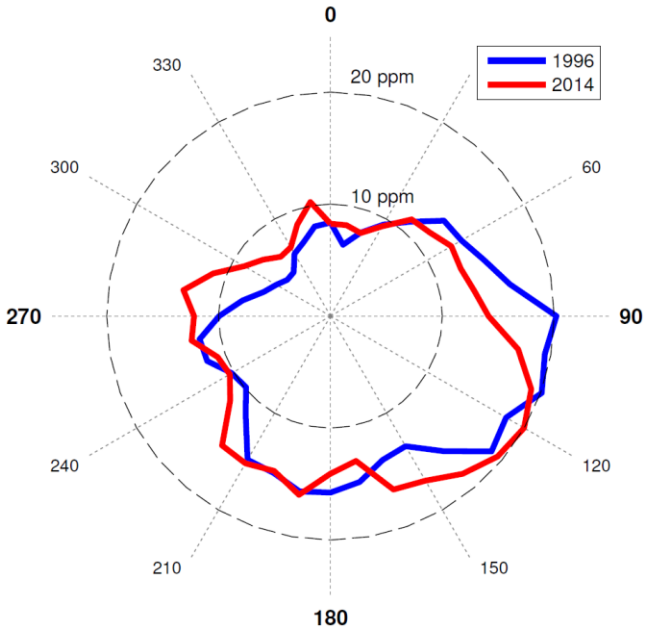

Figure 9: Wind rose of the mean excess CO_2 concentrations (concentration above background) measured at Cabauw at 200 m in 1996 (blue) and 2014 (red).

3.1.3 Emission indicator

Figure 10~~Figure 10~~ shows the Cabauw indicator for CO_2 per month. The seasonal variation shows a larger amplitude for the start of the measurements and mid 90's than now. The negative slope of the regression equation (-1×10^{-5}) suggests that the emissions of CO_2 in the Netherlands have decreased.

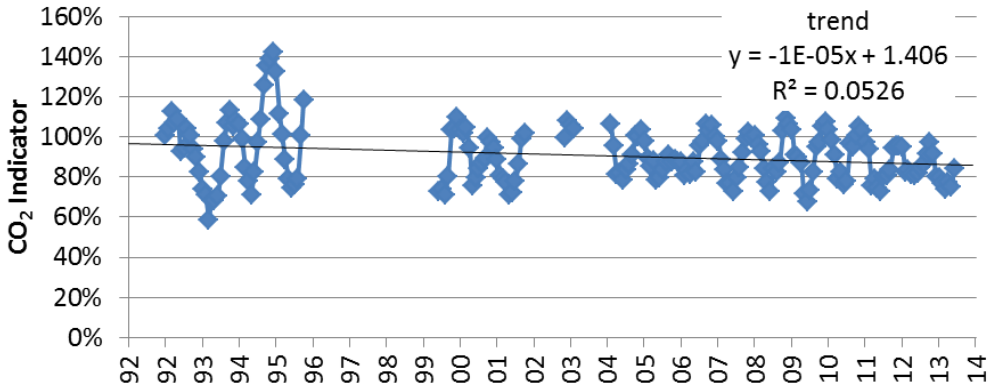

Figure 10: CO_2 emission indicator with a 6-monthly moving average calculated from the CO_2 concentrations measured at Cabauw at 200 m.

A comparison of the monthly indicators averaged per year with the emission registration numbers is shown in Figure 11~~Figure 11~~. This figure shows that the emission indicator fluctuates more strongly over the years than the CO_2 emission registration, with relatively low values in 1994 and 2000, and high values in 2004. In

contrast to the emission registration, the CO_2 indicator shows a decrease of 11% (for the years 2010 till 2012 compared to the years 1993, 1995 and 1996).

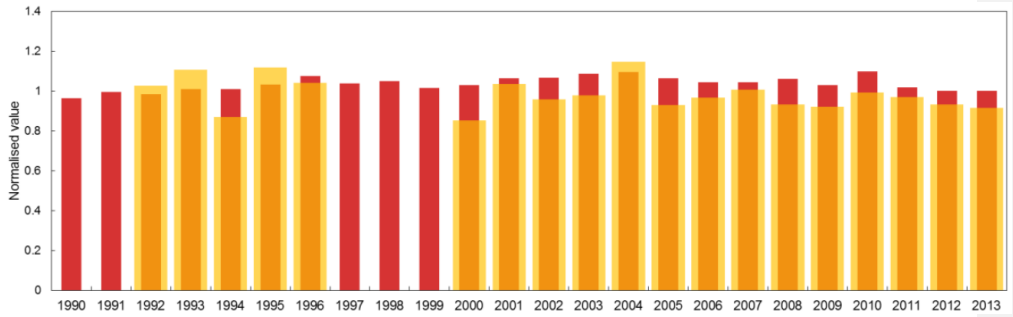

Figure 11: Yearly average of the CO_2 indicator (yellow) and the emission registration of CO_2 (red) both normalized.

Figure 12~~Figure 12~~ shows the results of the Cabauw indicator for CH_4. Both the indicator and the emission registration show a clear reduction in emissions. However, the 24% reduction estimate obtained from the indicator is smaller compared with the emission registration value of 37%.

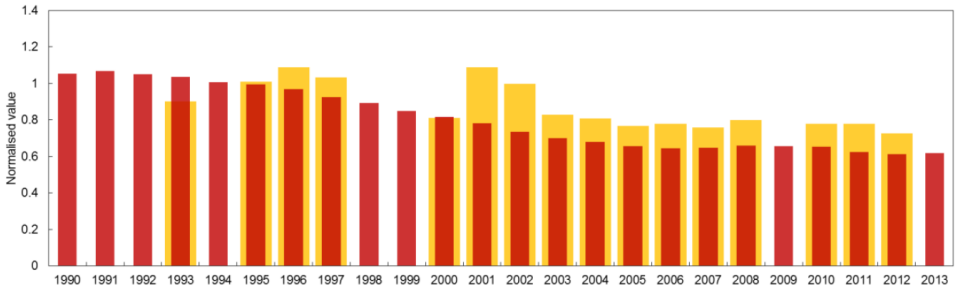

Figure 12: Yearly average of the CH_4 indicator (yellow) and the emission registration of CH_4 (red) both normalized.

The indicator corrected for the boundary-layer height for CO_2 is shown in Figure 13~~Figure 13~~. With this correction the reduction in emission is no longer visible (trend is 0%). This implies that over the years the boundary-layer height has increased, which leads to a lower difference between the 10th and the 90th percentile. The trend with the correction does correspond better with the emission registration.

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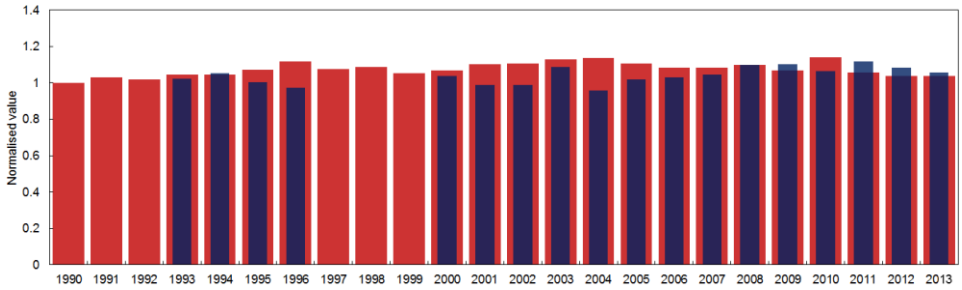

Figure 13: Yearly average of the CO_2 indicator corrected for the boundary-layer height (dark blue) and the emission registration of CO_2 (red) both normalized.

Figure 14~~**Figure** 14~~ shows the emission indicator corrected for the boundary-layer height for CH_4. The downward trend in emissions as reported by the emission registration is no longer observable in the Cabauw index. The Cabauw index in 2012 is more or less the same as that in mid 90s, which indicates that the downward trend observed in **Figure** 12~~**Figure** 12~~ are caused by an increase in the boundary-layer height.

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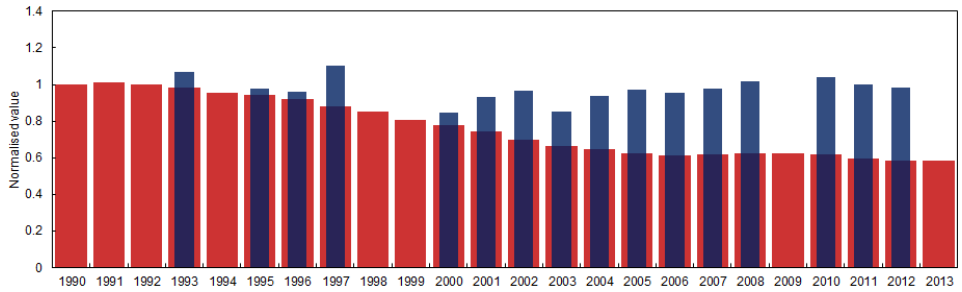

Figure 14: Yearly average of the CH_4 indicator corrected for the boundary-layer height (dark blue) and the emission registration of CH_4 (red) both normalized.

Note that the correction of the boundary-layer height uses ERA-interim data from the ECMWF model. Due to the long time series necessary this was the only dataset available. However, from November 2008 till December 2011 a LIDAR instrument was measuring the boundary-layer height at Cabauw. **Figure** 15~~**Figure** 15~~ shows the difference between the boundary-layer height of the ERA-interim dataset and the LIDAR measurements over this time period. There are clear discrepancies between the two datasets, especially in summer with differences up to 2000 m.

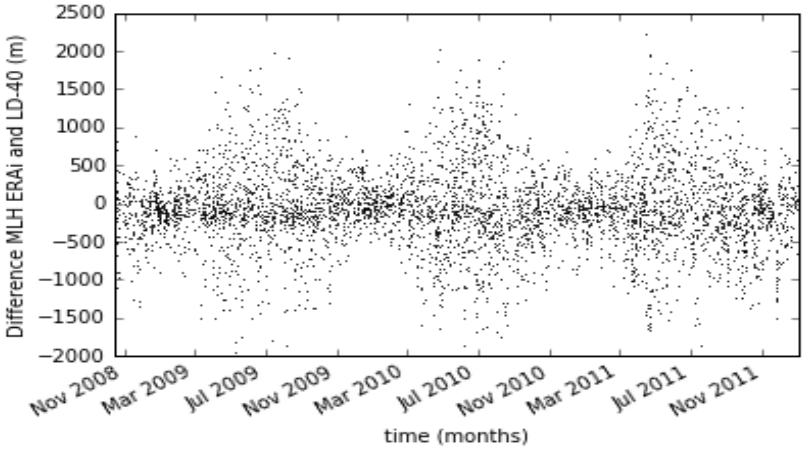

Figure 15: Time series of the difference in boundary-layer height between the ERA-interim data and the LIDAR – measurements at Cabauw.

Also for other components the emissions indicators can be calculated (e.g., CO, N_2O, and SF_6). However, for these components the time series of the Cabauw data are shorter (starting in 2004) than those of CO_2 and CH_4 (starting at the end of 1992).

3.1.4 Spectronus performance

The Spectronus started measuring at Cabauw at the beginning of 2015. Before the measurements at Cabauw started, the performance of the Spectronus was investigated and improved at ECN in Petten (2013-2014). The following improvements were made (also shown in **Figure** 16~~**Figure** 16~~):

- Cell interior polished.
- Gold plating of the metal cell.
- High precision temperature sensors (1 mK) added at three locations in cell.
- Local heating pads added to control the cell temperature.
- External program created for control of complex cycles, which triggers samples, work targets, and calibration measurements.

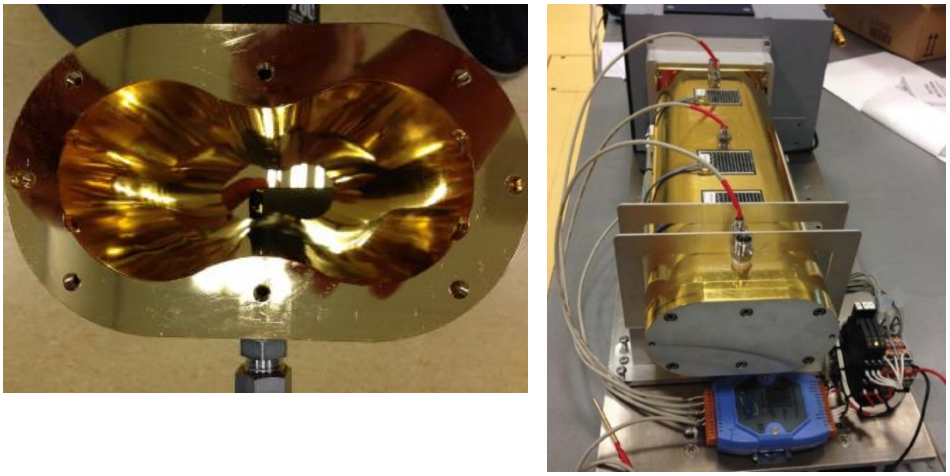

Figure 16: Improvements made on the Spectronus with on the left the inside of the gold plated cell and on the right the outside with the temperature sensors and local heating pads visible.

Instrument testing with Alan Variance technique.

Testing the performance of the instrument is done using a so called Alan variance test. This test uses a measurement with the instrument for a relatively long time period, for example a few hours on a gas flask, with one well defined concentration level. The test looks how the standard deviation of the measurement changes with longer and longer measurement intervals, from 1 measurement to 10-100 or 100 measurements. At the start averaging more data at first cancels out the noise, providing a better defined concentration level. When averaging over an episode that is too long however, the standard deviation starts to increase again due to drift in the measurement system. The lowest value found is the best performance option for the system under investigation.

The improvements resulted in lower Allan variances (see text box above) in static mode for all the measured gasses (CO_2, CH_4, N_2O, CO, and $^{13}CO_2$), see **Table 2**~~**Table 2**~~. For CO also the drift per day decreased to 0.04 ppb.

Table 2: Allan Variance, precision and drift of the different species measured by the Spectronus after the improvements.

Species	Allan Var				Precision	Drift	Unit
	flow 1 min	flow 5 min	static 1 min	static 5 min	stdev 3 days	per day	
CO2	0.018	0.007	0.018	0.014	0.031	0.021	ppm
13CO2	0.08	0.04	0.03	0.02	0.07	0.03	permille
CH4	0.18	0.10	0.20	0.10	0.18	0.11	ppb
N2O	0.15	0.07	0.12	0.05	0.08	0.009	ppb
CO	0.25	0.12	0.20	0.07	0.14	0.04	ppb

A disadvantage of the Spectronus compared to other measurement devices is that the measurement cell is relatively large (for the one deployed at Cabauw 2.5 L). The calibration cycles applied nowadays for gas measurements at tall towers are repeated daily. However, applying that to a Spectronus the calibration cylinders would already empty after just over 3 years. For long term measurements it is undesirable to use calibration cylinders so shortly, thus a new calibration scheme is necessary to ensure continuity in the measurements. Besides the calibration, also working standards and target measurements are applied. An optimum between the amount of calibrations, working standards, and target measurements applied and the amount of gas used to do the measurements. At the moment the following scheme is applied:

- Calibration cycle → once every 5 days
- Work target 1 → every 6 hour
- Work target 2→ every day

Careful data analyses and testing of different schemes are necessary to find the optimal setting for the calibration scheme. In the coming year, these analyses and testing will be done to find the optimal settings.

Figure 17~~Figure 17~~ shows an example of the time series of the Spectronus CO_2 measurements.

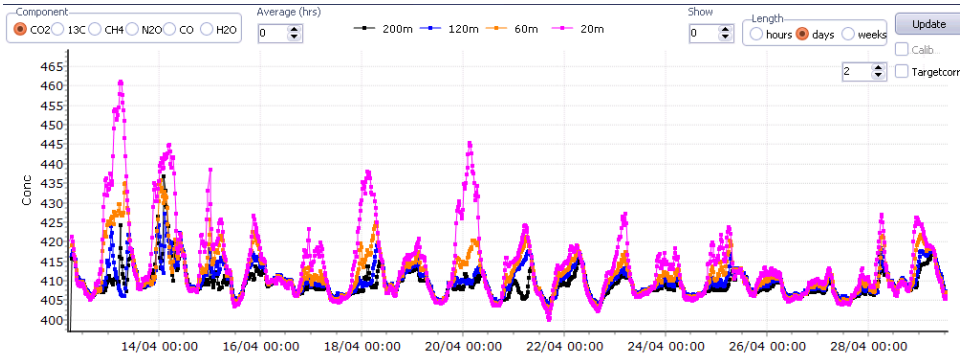

Figure 17: Example of the time series of CO_2 measured by the Spectronus, with in pink the 20 m level, orange 60 m level, blue 120 m level, and in black the 200 m level.

3.2 Data use

3.2.1 International data availability

The data collected at the Cabauw tower are widely used by scientists in the Netherlands and abroad. There are several reasons why scientists use the Cabauw data:

- Long time series
- Multiple gas species
- Multiple heights
- Large signal from sources

One of the reasons is the long time series; the first CO_2 measurements started in 1992 and therefore provide one of the longest continuous measurement series in the world. Over the years, with the development in measurements techniques the CO_2 measurements became more accurate. Furthermore, also multiple components were measured: CH_4, N_2O, CO, and SF_6. As of 2000, after a renovation of the Cabauw tower, the measurements are taken at 4 heights (200 m, 120 m, 60 m, and 20 m), another reason for scientists to use Cabauw data. Due to its location in an area with different sources, the Cabauw dataset covers both clean air and clearly polluted air masses. The measurements are therefore essential to improve model calculations in and downwind of, source areas.

The collected Cabauw data are spread to scientists by different means. First of all, there are international collaborations like the InGOS and ICOS projects. There are also other

initiatives like Observation Package (ObsPack) by the National Oceanic and Atmospheric Administration (NOAA), which make data available to scientists. Second of all, there are scientists that directly ask us to supply data to them. This distribution of data has led to scientific advancement, which can be seen by the high amount of scientific publications that use the Cabauw data collected by ECN (see Appendix 1 for a list of the publications).

ObsPack

The collected CO_2 data at Cabauw are submitted to ObsPack. The main aim of ObsPack is to stimulate and support carbon cycle modelling studies. The data are freely available to the scientific community under a fair use statement, which states that an agreement has to be reached between data provider and data user to insure that an appropriate level of acknowledgements is reached.

The ObsPack data were made available at the end of July 2015. In total the data have been downloaded 97 times in 2015, by people around the globe (see **Figure** 18~~**Figure 18**~~).

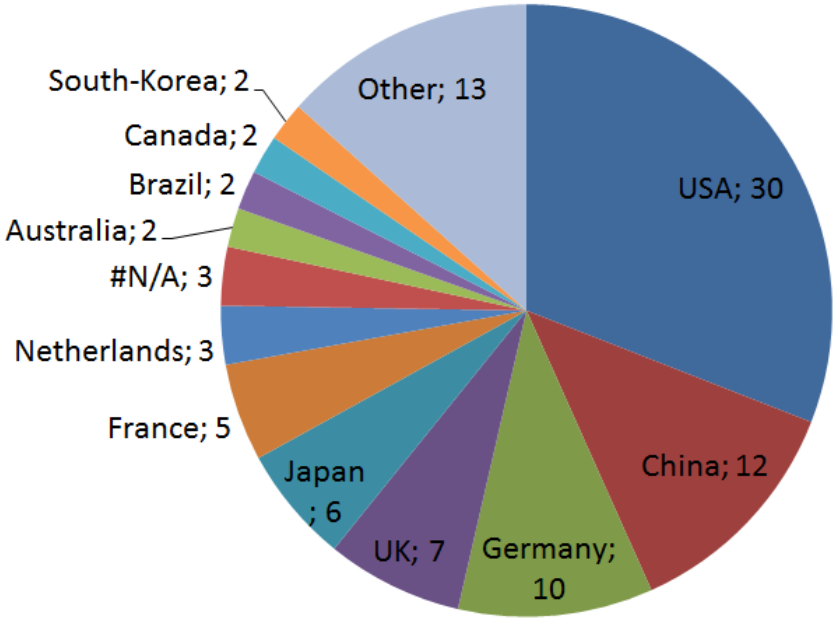

Figure 18: Amount of downloads of ObsPack data per country.

3.2.2 InGOS inverse modelling results (Source: InGOS 3rd period report)

Inverse models use a combination of measurements and a transport model to calculate emission maps. During the InGOS project this approach was implemented to derive the emission maps of GHG's over Europe.

The modelled emission map of CH_4 is shown in **Figure** 19~~**Figure 19**~~ using seven different inverse models (and the a priori emissions as applied in TM5-4DVAR). Similar spatial emissions patterns are visible on small scales (<200 km), due to the fact that the same a

priori emission map is used. On larger regional scale moderate changes of emissions were calculated compared to the a priori emissions.
The total emission over Europe (EU-28) for 2006-2012 as calculated by the inverse model is 28.4 ± 6.4 Tg CH_4 yr^{-1}. The reported CH_4 emissions of the bottom-up inventories to UNFCCC are lower with 19.0 – 20.9 Tg CH_4 yr^{-1}. However, the reported uncertainties of the total emissions of CH_4 per country are in the order of 20-30%. A possible explanation for this difference between the top-down (inverse models) and the bottom up inventories are the natural sources. For the bottom-up inventories the contribution of natural courses were assumed to be relatively small (except for Scandinavia).

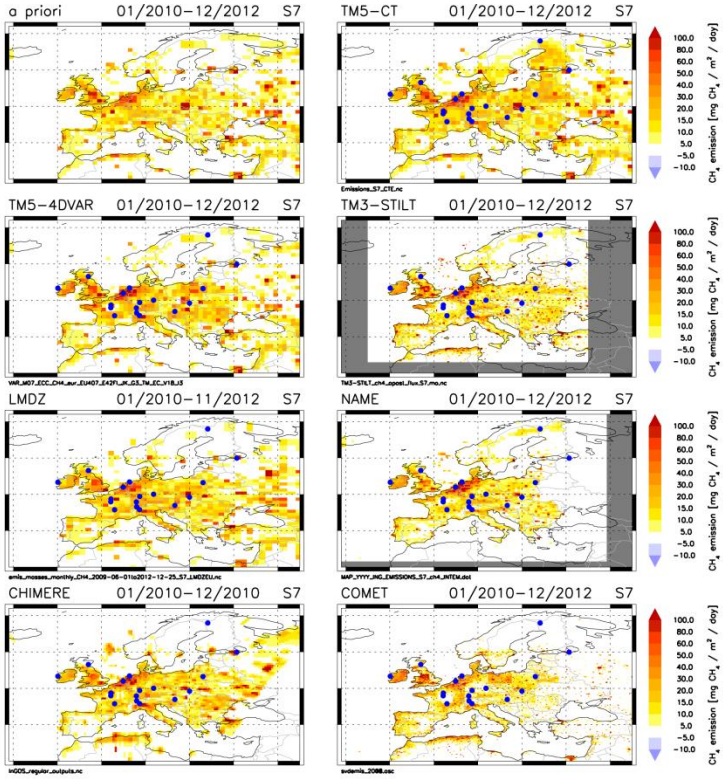

Figure 19: Maps of the European CH_4 emissions (average of the years 2010-2012) for different inverse models. The a priori emissions as applied in TM5-4DVAR are shown in the upper left panel. (Source: InGOS final report).

For N_2O four different inverse models were compared during the InGOS project. The calculated N_2O emission maps are shown in **Figure** 20~~**Figure** 20~~ (again as well as the a priori emission map as in TM5-4DVAR). The maps show higher emissions in North-western Europe than the prior for the TM5-4DVAR, TM3-STILT, and LMDZ. For the NAME model the emission map is relatively close to the a priori emission map.
The inverse models calculate a total emission of N_2O of 1.41 ± 0.54 Tg N_2O yr^{-1} over Europe (EU-28). The bottom-up inventories reported to the UNFCCC show a decreasing trend in total anthropogenic N_2O from 1.23 Tg N_2O yr^{-1} in 2006 to 1.08 Tg N_2O yr^{-1} in 2012. The estimated natural soil emissions of N_2O over Europe are very small (~0.1 Tg N_2O yr^{-1}). Note that the reported uncertainty of the bottom-up inventories is very large for many countries more than 100%. These large uncertainties are mainly caused by the

emissions from the agricultural source which exhibit strong spatial and temporal variability making upscaling very difficult.

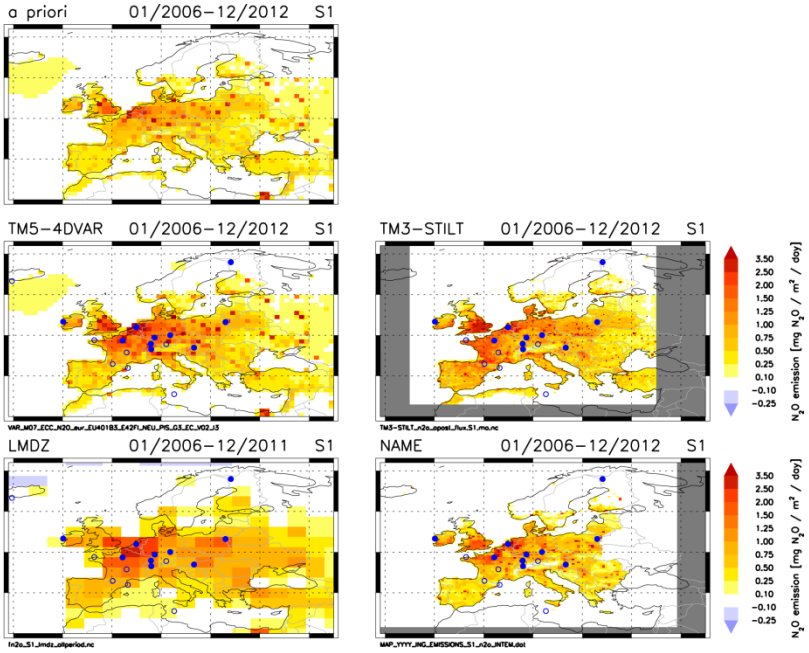

Figure 20: Maps of the European N_2O emissions (average of the years 2006-2012) for different inverse models. The a priori emissions as applied in TM5-4DVAR are shown in the upper left panel. (Source InGOS final report).

3.2.3 InGOS Cabauw inter-comparison campaign

The gas exchange between the soil and atmosphere can be estimated by flux measurements. With that, both a sink or source at the ground level can be determined. A common method to calculate fluxes (known as the eddy covariance method), is by combining high frequency measurements of a quantity (can be GHG-concentrations, but also other quantities like temperature) with 3D wind data.

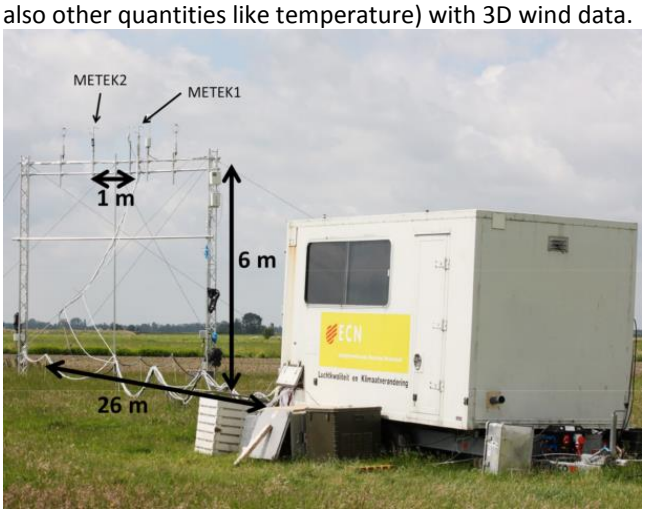

Figure 21: Experimental setup during the inter comparison campaign.

During the InGOS project an inter comparison experiment with eight fast-response methane analysers was undertaken in June 2012 at Cabauw (see Figure 21~~Figure 21~~). The analysers were tested on data coverage and quality, amount of noise, magnitude on simplicity of different corrections, and the agreement of the fluxes. The different instruments compare very well to one another, most differences occur in data coverage and the amount of noise. Especially, the water correction needed was difficult to do for systems that did not measure the water concentration itself. In the end all analysers (except G1301-f and LI-7700 due to the low amount of data coverage) showed the same cumulative flux pattern within ±10% over a period of 13 days (see Figure 22~~Figure 22~~). For more information on the campaign see Peltola et al. (2014).

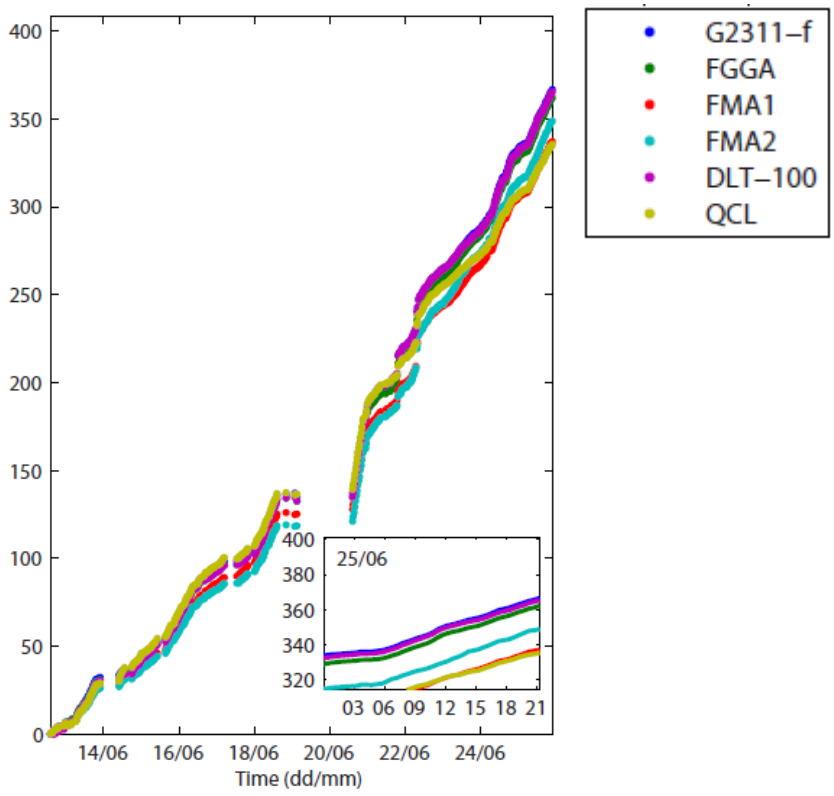

Figure 22: Cumulative sums of the CH_4 fluxes during the inter-comparison campaign from 12 to 25 June after the H_2O corrections (Peltola et al., 2014).

The important lesson of this campaign was that the methane measurement systems available now are able to quantify the net methane emission and they all agree within 10%. The other conclusion was that the site to site difference can be much bigger: three different sites in a 5x5 km apart showed differences up to 100%.

4
Conclusions

With support of the Ministry of Environment and Infrastructure the Cabauw GHG measurements were continued in January 2015. This report documents the work achieved in 2015 in the initiated project and the associated work achieved for the InGOS EU project which ended March 2016.

The aim of the greenhouse gas measurements at the Cabauw tower is:

- to keep track of GHG emissions in the Netherlands and beyond;
- to understand and evaluate the effect of the changing climate on natural GHG exchange;
- to provide measured evidence to support decision making in the climate change dossier.

The Cabauw dataset is considered special worldwide because of:

- the length of the time series (22 years);
- the availability of multiple measurement heights;
- The Cabauw geographical position in the centre of important European source area of GHG's.

The Cabauw data is used worldwide for atmospheric transport model developments and improvements, for example through Obspack. The advantages mentioned above are an important reason for this. The other reason is the high data quality and the fact that the data is internationally well embedded through NOAA and European intercalibration schemes.
Keeping the data quality up to par with the international community requires a constant effort. At the beginning of 2015, a fast Fourier transform infrared spectrometer has been added to the Cabauw measurement setup. This improves the data quality of the CO_2, CH_4, CO, and N_2O measurements. Further, this measurement device is also able to measure ^{13}C.
In the 2016, the final setup concerning the calibration cycle will be determined for this measurement device. With the addition of the fast Fourier transform infrared spectrometer, the old GC system has been removed from the measurement setup, due to its high maintenance costs and poorer quality of the data. This choice and the lack of funding means that unfortunately SF_6 is no longer measured at the Cabauw site.

The Cabauw GHG measurement can be used for different applications. The wind roses figures give a first indication of the source areas around Cabauw. The high peak concentrations seem to have decreased from 1992 to 2014 in the south east. However, overall the mean excess CO_2 concentration (the level above the background concentrations) per wind sector stayed more or less the same for 1996 and 2014, which indicates that on average the CO_2 emissions did not change severely.

The new concept of the Cabauw indicator is a step further than wind roses. It can provide trend information on the emissions level of greenhouse gases over the Netherlands. The first preliminary results for CH_4 seemed to compare fairly well with the emission registration. However, when the influence of the boundary-layer height of the atmosphere is taken into account the emissions of CH_4 does not decrease as suggested by the emission registration.
For CO_2 the Cabauw index and the emission registration agree well with one another when the boundary-layer height is taken into account (both show no significant decreasing or increasing trend).

With this the conclusions for the new proposed Cabauw emission indicator are:

- The Cabauw indicator confirms the trend in the Dutch CO_2 emission (which is assumed to have only a 3% uncertainty in the emission registration estimates).
- The Cabauw indicator does not support the emission reduction for CH_4 and suggests that there is hardly any decrease. Which is in line with the results of the inverse modelling calculations that indicate the Benelux emission level for methane might be 40% above the estimated level (not shown in this report, research still in progress).
- The indicator calculations corrected for the boundary-layer height revealed that as a result of an increase in boundary-layer height the concentration levels have increased less prominent over the last 20 years which is also important information for the evaluation of air pollution data.

Recommendations:

- Evaluate the importance of the boundary-layer height with KNMI.
- Evaluate other single station emission evaluation techniques to check the Cabauw indicator calculations for internal consistency.
- Restart SF_6 measurements and start halocarbon measurements.
- Continue the Cabauw GHG monitoring.

Boundary-layer height:
In order to correct the Cabauw indicator for changes in the boundary-layer height the ERA-interim dataset (based on re-analysed of the ECMWF weather maps) was used which showed discrepancies compared to the measured boundary-layer height by the LIDAR. In order to get a better grip at the influence of the boundary layer-height on the Cabauw indicator a better estimate of the boundary-layer height is necessary. Colleagues at the KNMI (the Royal Netherlands Meteorological Institute) can estimate the boundary-layer height from the historical meteorological data gathered at Cabauw. Thus, by working together we can understand the influence of the boundary-layer height on the emission level estimates better.

Test other single station techniques:
Another option to estimate the emissions of greenhouse gases from the observation is by using radon as a reference tracer. Radon is measured at two heights at Cabauw (20 and 200 m). The surface emissions of radon are available from European soil flux map (see for example those of Szegvary, 2007). From these surface emissions and the difference in concentration between a baseline and concentration during an event for radon and another tracer, the emission of the tracer can be calculated (among others van der Laan et al. 2009). So far, this method to obtain emissions from radon measurements has not been applied to the Cabauw data.

Restart SF_6 and start Halocarbons measurements:
The impact of SF_6 and Halocarbons on the radiative balance of the atmosphere is increasing rapidly. Already the combined effect of this suite of different gasses used for foam making, insulation and fire extinguishing is larger than the impact of N_2O, the bronze medal winner after CO_2 and CH_4 in the GHG impact contest thus far. The measurement of these gasses now takes place mainly at semi background stations. The Cabauw tower again is amidst several source areas in the Benelux and Germany. Since all emission reduction efforts here are only covenant, based independent measured evidence that these agreed promises are indeed lived up to are necessary. These measurements are of special importance in the world wide discussion which country/continent is in fact the main source of many of these components.

Continue measurements:
The funding from the Ministry of I&M and InGOS enabled ECN to continue Cabauw GHG operations and calibrate, quality check and evaluate the data collected from 2011 till 2015.

From the text above it is clear that measurements of greenhouse gases are essential to assess their emissions and the impact of a changing climate on the natural emissions. In order to reach the goal set in Paris at the climate top of a maximum increase of global temperature 2 degrees, drastic measures in reducing emission levels are necessary. The measurements serve as a way to ensure and show to the public that measures taken have the desired effect (or not). Furthermore, they also are essential input for researchers to understand the complex atmospheric system of sources and sinks of greenhouse gases.

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Appendix A. List of scientific publications that use the ECN Cabauw GHG data

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