



# Mitigation

## An assessment of the potential for atmospheric emission verification in The Netherlands

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# Contents

Summary in Dutch	5
Summary	5
Extended summary	6
Preface	6
1. The need for atmospheric emission verification	7
Introduction	7
Understanding greenhouse gas dynamics	7
The ideal monitoring system	13
Only the atmosphere matters	14
2. The monitoring system for greenhouse gas emission verification	15
The current network vs the ideal network	15
In situ monitoring of greenhouse gas concentrations	15
Remotely sensed greenhouse gas concentrations	17
Direct observations of regional fluxes	20
Monitoring of Atmospheric Boundary Layer dynamics	23
3. An inverse modelling system for greenhouse gas emission verification	27
Inverse modelling and transport model performance	27
Inverse modelling algorithms	37
Continental inversions	39
Regional scale inversions: towards estimation of Dutch GHG emissions	42
4. Can we verify reported emissions ?	44
Conclusions present study	44
Current status emission verification	46
Outlook for Verification	49
References	52



# Summary



## Summary in Dutch

Doel van het Klimaat voor Ruimte project ME2: *Integrated observations and modelling of Greenhouse Gas budgets at the national level in the Netherlands* was het ontwikkelen van een systeem voor het kwantificeren van het broeikasgasbudget op landelijke en regionale schaal. Het ME2 consortium heeft een 'protocol' ontwikkeld om een referentieschatting te maken ten behoeve van de verificatie van nationale emissies. Daarmee is het op termijn mogelijk de nauwkeurigheid en geloofwaardigheid van aan UNFCCC en Kyoto gerapporteerde emissies, en reducties daarvan, te verifiëren.

Het project heeft een meetnetwerk voor broeikasgasconcentraties en grenslaag eigenschappen geoperationaliseerd dat verificatie tot op zekere hoogte mogelijk maakt. Implementaties en extensies zijn ontwikkeld van hoge resolutie emissie en transport modellen voor CO<sub>2</sub> en CH<sub>4</sub> (RAMS, WRF, COMET, TM5) en gevalideerd met metingen uit het netwerk als referentie. Met verschillende inversie methoden, van data tot model gedreven, zijn emissieschattingen gemaakt. De data gedreven methoden kunnen schattingen maken voor alle drie de broeikasgassen voor NL als geheel en zijn representatief voor meerdere jaren. Met de meer model gedreven inversies zijn meer ruimtelijk en temporeel gedistribueerde schattingen te maken. De nationale inversieschattingen lijken voor vooral methaan en lachgas hoger te zijn dan op basis van de officiële rapportages, maar de onzekerheden zijn (nog) groot (25-30%).

Het project heeft veel gepubliceerd, in druk en bij wetenschappelijke congressen, maar ook gericht op beleid zoals bij o.a. CoP15, VROM. Continuering wordt gezocht in het ICOS project, maar een substantiële Nederlandse bijdrage aan dit Europese initiatief is december 2011 nog niet zeker gesteld.

## Summary

The *Climate Changes Spatial Planning (CcSP)* project ME2: *Integrated observations and modelling of Greenhouse Gas budgets at the national level in the Netherlands* aimed to “develop an advanced GHG information system - consisting of a comprehensive set of monitoring systems, combined with a complementary suite of 3D models – that is able to quantify the magnitude, trends and associated uncertainties of the biogenic and anthropogenic greenhouse gas budgets at high spatial and temporal resolutions”. While doing so, a protocol has been developed to provide an independent reference estimate for the verification of national emissions, and reductions thereof, as reported by the parties in the UNFCCC and Kyoto framework.

The project operationalized a sensor network (tower based and airborne) to monitor GHG concentrations and boundary layer dynamics. High resolution emission and transport models (RAMS, WRF, COMET, TM5) have been developed for CO<sub>2</sub> and CH<sub>4</sub> and validated against data from the monitoring network. Both data and model based inversion methods have been developed and used to produce emission estimates. Data based methods allowed estimates at national scale representing multi-annual budgets, while model based estimates allowed spatially explicit, seasonal estimates to be made. Resulting national estimates suggest that emissions for N<sub>2</sub>O and CH<sub>4</sub> may be higher than reported, though typical uncertainties are high.

The consortium published extensively in peer reviewed literature, but also addressed policy community by presenting at e.g. COP15 and at national ministries. Continuity of this research is sought through the European ICOS initiative, but national support has not been confirmed yet (December 2011).



## Extended summary

For an extended summary we refer to pages 44-51.



## Preface

This report makes an assessment of the potential for atmospheric emission verification in the Netherlands. As such it is the final product of the *Climate Changes Spatial Planning (CcSP) project ME2: Integrated observations and modelling of Greenhouse Gas budgets at the national level in the Netherlands*. This project aimed to “develop an advanced GHG information system – consisting of a comprehensive set of monitoring systems, combined with a complementary suite of 3D models – that is able to quantify the magnitude, trends and associated uncertainties of the biogenic and anthropogenic greenhouse gas budgets at high spatial and temporal resolutions. While doing so we will develop a protocol to provide an independent reference estimate for the verification of national emissions reported by the parties in the UNFCCC and Kyoto framework”.

This project, co-funded from a number of other national and European funded sources, allowed the consortium to further develop in close collaboration a number of related research lines approaching the problem from several perspectives. These include monitoring of both high precision GHG *concentrations* at tall towers and of regional turbulent GHG *fluxes* from the same high towers and from aircraft. It included the monitoring of important atmospheric transport characteristics like spatially distributed boundary layer height dynamics and its mixing efficiency.

The following report does not intend to give an administrative account of project activities and deliverables. Rather it synthesises the scientific results of the project in the context of the state-of-art of the respective research fields (Chap 2 and 3) and describing the societal relevance and potential of the results (Chap 1 and 4). In the reference section we identify those publications that have been made possible through contributions of ME2, distinguishing them from publications from the wider community.

We intend to convey a clear message as to where we are right now in terms of scientific potential and limitations for independent verification of bottom up, basically self-reported emissions. We aim to give a blue print for the necessary ingredients of a system that can perhaps provide in the near future such independent emission estimates in a more operational way, ingredients that include monitoring hardware, modelling software and human resources (and continuity in funding).The



emissions of GHGs in the Netherlands are among the highest worldwide when expressed per unit area due to the high population density, the high economic development based on fossil fuel use and the large agricultural activity focused on production of meat and dairy products. These emissions are concentrated in hotspots in the urban areas but are also distributed over the countryside for the agricultural sector.

## 1. The need for atmospheric emission verification

### Introduction

Our climate is changing fast and this is primarily caused by increased concentrations of greenhouse gases, caused by emissions into the atmosphere by mankind. Atmospheric trace gases like the naturally occurring carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O), but also others like the synthetic fluorinated gases absorb heat and thereby change the radiation balance of the earth. This increased radiative forcing of climate is beyond dispute. Although the concentrations of natural greenhouse gases (GHGs) exhibited strong cyclic variations in the distant past as a result of the non-linear response of the biosphere to other forcings (e.g. solar precession), current changes through human influences are unprecedented in rate and size. The non-linear behaviour of the Earth's climate system is due to strong, but poorly understood feedbacks, one of these being the feedback between climate and the production and decomposition of organic material. However, industrial emissions of fossilized carbon and augmented turnover of carbon and nitrogen associated with increasing agricultural production have driven GHG concentrations far above levels found at any period in the past million years.

Managing climate change has become one of the big challenges mankind will have to address in the coming decades. Adaptation to a level of climate change we are committed to as a consequence of already realized GHG increases, must be accompanied by mitigation of further GHG concentration/emissions. Mitigation of GHG emissions requires understanding of the complex interactions between the climate system, natural GHG dynamics and human perturbations. These need to be adequately represented in climate models in order to be able to produce realistic projections into the future. But they also need to be known in order to be able to realize an (economically) optimal mix of mitigation activities. Both scientific understanding and management tools are in need of proper data on GHG dynamics and its drivers. For this to be achieved we will ultimately need process models both to integrate understanding of past realizations and to also allow projections into the future. Those two needs largely overlap, but also differences exist in e.g. the acceptable levels of uncertainty, or the desired geographical focus and level of detail

### Understanding greenhouse gas dynamics

Understanding variability and trends in greenhouse gas concentrations starts with observed data. The more than 5 decades long measurement series of atmospheric CO<sub>2</sub> concentrations started by Keeling on Mauna Loa (e.g. (Keeling, Whorf et al. 1995) has been pivotal in raising awareness of human interference with the atmosphere. It provides a canonical view on seasonal and intra-annual, mostly natural CO<sub>2</sub> variations (Rodenbeck, Houweling et al. 2003) that are superimposed

on a steadily increasing concentration trend due to anthropogenic emissions. It is the continuity of such records that helps us to put events in their proper contexts, such as dip in global CO<sub>2</sub> uptake after the 1991 volcanic eruption of Mount Pinatubo, or the peak due to increased fire emissions in Indonesia in 1997/1998 following a very strong El Niño, or the 2003 European drought. These data also show that only 45% of human emissions of CO<sub>2</sub> remain in the atmosphere (le Quere, 2010), the rest is taken up by oceans and terrestrial vegetation, a discount (negative feedback) we are not sure we can rely on in the future (IPCC 2007), as both current trends and model predictions suggest that the ocean and land uptake may decrease (Heimann and Reichstein 2009; Rockström, Steffen et al. 2009) leading to an increase of the so-called airborne fraction. These continuous records also show that from 2005-2008 actual global emissions exceeded one of the highest SRES scenarios (A1FI) made by the IPCC as recently as 2001 (Canadell, Le Quere et al. 2007; Le Quere, Raupach et al. 2009), after which global emissions stabilised due to the economic crisis (Olivier and Peters 2010). Likewise, the GHG records reconstructed for the past 800000 years from ice cores (Lüthi, Le Floch et al. 2008) have shown us the natural limits between which GHGs varied and has forced upon us the notion that the current rate of increase is truly unprecedented. It also increasingly provides constraints on the strength of natural feedbacks between climate and GHG dynamics.

Basically, three classes of measurement approaches to establish GHG budgets exist: 1) computing fluxes from differences in carbon stocks, 2) directly measuring fluxes between the earth surface and the atmosphere, and 3) so-called atmospheric inversion techniques. In the following paragraphs we will briefly discuss the basics of each. Somewhat more detail will be given on the first two approaches than on the third. The latter approach is the main focus of this report and will be discussed extensively in chapters 2 and 3.

From a long term perspective we are mostly interested in earth system states, i.e. the (equilibrium) stocks of carbon in oceans (inorganic and biotic) and on land (living and dead biomass and soil stores) and their changes. It is important to note that for most carbon stores 'slow in, fast out' applies. Increasingly longer timescales are associated with the storage of carbon in living biomass (wood), soil carbon or fossil fuels respectively. In each case human extraction of the same carbon and release to the atmosphere is (many) orders of magnitude faster. Fluxes between the compartments can be estimated from changes in stocks, but involves differencing large numbers estimated at long intervals. Dense, but technically relatively simple sampling is required to ensure that the uncertainty in the subsequent stock assessments is smaller than the change we are interested in. E.g. European forest inventories rely on repeated sampling at 5-10yr intervals of 100000 plots check.

At the global scale and considering only above ground carbon, the carbon in woody biomass (forest) is accounted from forest area (mostly remote sensing based) and average biomass per hectare (ground based biomass plots). Thus, also changes in forest area can be converted to a flux. Global deforestation rates and fluxes for the 1980's have been estimated at 7.8 Mha/yr representing a flux of -1.4 (+/-0.9) GtC/yr and for the 1990's 8.9Mha/yr representing a flux of 1.6 GtC/yr, almost all in the tropics (IPCC 2007). In Europe and North America relatively young forests act as a sink. In Europe the forest sink strength based on inventory data was estimated at 236-542 MtonC/yr (Janssens, Freibauer et al. 2003) recently updated to 220 +/-43 MtonC/yr (Schulze, Luyssaert et al. 2009). These numbers serve as an illustration of the uncertainties typically involved. For more complete assessments at e.g. the European scale, see these publications and also (Janssens, Freibauer et al. 2005) or (Nabuurs, Thurig et al. 2008).

In the Netherlands UNFCCC reporting of emissions from land use and land use change (LULUCF) is also solely based on inventory data. An annual land cover transition matrix derived from detailed land use maps provides conversion rates from forest to non-forest lands and vice versa. Carbon



stock changes in the Netherlands are thus calculated to be a net source of CO<sub>2</sub>, amounting in 2007 to 2.5 Mton/yr. This number is the sum of increased stocks in new (-0.6 Mton) and existing (-2.2 Mton) forests, and decreased stocks in cultivated (grassland) and drained peat soils (4.2 Mton/yr), which exceeds the sequestration of carbon in forestry. Croplands are assumed to have stable, i.e. non changing carbon stocks in their soils (van der Maas, Coenen et al. 2009). In a separate study uncertainties of these inventory based fluxes were estimated (Olivier, Brandes et al. 2009). According to this work the forest sequestration estimated from stock changes has an uncertainty of 64%, the emissions from peat oxidation are known only within 56%. The total LULUCF emission uncertainty is estimated to be 103%. Though these are high-end estimates, especially for the 'activity data' - i.e. land use map data, it is clear that even in a data rich country like the Netherlands uncertainties in biogenic sources or sinks from inventory data are large and of the same order of magnitude as the fluxes themselves.

The basic carbon stock estimates are continually being improved. Recent efforts focused on e.g. the spatial distribution of soil organic matter (SOC) and forest floor carbon stock (FFC). Tree species, age of the stand as well as management proved to be a important sources of variability on both the SOC as the FFC (Schulp, Nabuurs et al. 2008). Likewise, the influence of land use history on the carbon stock has also been studied (Schulp and Veldkamp 2008). The historical land use proved to explain a larger part of the variability than the present land use,  $r^2 = 0.20$  for 1850 land use and  $r^2 = 0.14$  still for 1780 land use as compared to against  $r^2 = 0.02$  for present day land use (the remaining variability explained by soil texture and groundwater). Including this land use history in a national-scale inventory of SOC and FFC stocks improved the SOC and FFC stocks by 5-10%. Increasing the sample density did not decrease the error of SOC in agricultural lands (Schulp, Verburg et al. SUBM). Area and site based studies are important for processes and large scale accounting methods. Development of mitigation options, however, is better served with studies at appropriate management levels. An example in the current context is the farm level. Model based studies on the influence of dairy farm management on Dutch soil carbon stocks (and integral GHG emissions) showed that grassland productivity and the amount of manure application may change its soil carbon stocks by 5-10% (van Evert and Verhagen in prep). See also Kvr Report (KvR056/12).

Much more useful for understanding the dynamics at shorter time scales and for relating rates of changes to environmental drivers is the direct measurement of fluxes. However, gross fluxes are much larger than the net fluxes that matter. Photosynthetic and respiratory fluxes are one or two orders of magnitude larger than net ecosystem or net biome exchange respectively. Technically complicated monitoring techniques are to be maintained over long periods, in order to sample a wide enough range of variations in drivers to provide confidence in the models we construct from these.

From flux tower data the forest sink in Europe was first estimated at 312 MtC/yr recently updated to 204 MtC/yr (Schulze, Luysaert et al. 2009). Uncertainties for these kind of estimates is in the order of 20%. In the Netherlands, as part of a sister project, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O fluxes have been monitored for at least a full seasonal cycle on many sites, representative for grasslands, various crops and forest. These showed considerable variability with classes, e.g. for grasslands CO<sub>2</sub> (Jacobs, Jacobs et al. 2007), N<sub>2</sub>O (van Beek, Pleijter et al. 2009) or CH<sub>4</sub> (Schrier-Uijl, Veenendaal et al. 2008). These were scaled up to the national level for CO<sub>2</sub> by (Garcia-Quijano, Tolk et al. 2011 (subm) who showed a considerable lengthening of the growing season in recent years associated with a more than doubled annual uptake. For a more extensive discussion of flux observation in the Netherlands see Kvr report no[insert report numer ME1].

The third approach, and the one we will focus on in this manuscript, makes use of the fact that the atmosphere is a fast but imperfect mixer of spatially and temporally varying surface fluxes. The distribution of regional fluxes over land and oceans can be retrieved using observations of gradients of atmospheric GHG concentrations that are unravelled through atmospheric transport models. To retrieve the GHG budget for any domain, one needs to characterize the inflow of GHGs, the outflow of GHGs, and the net difference between the two amounts to the total exchange (sources and sinks) between the atmosphere and the other GHG or carbon reservoirs. This so-called ‘top-down’ or atmospheric inversion approach was originally developed for global to continental scales (Tans, Bawkin et al. 1996), but in principle can also be applied at much smaller scales.

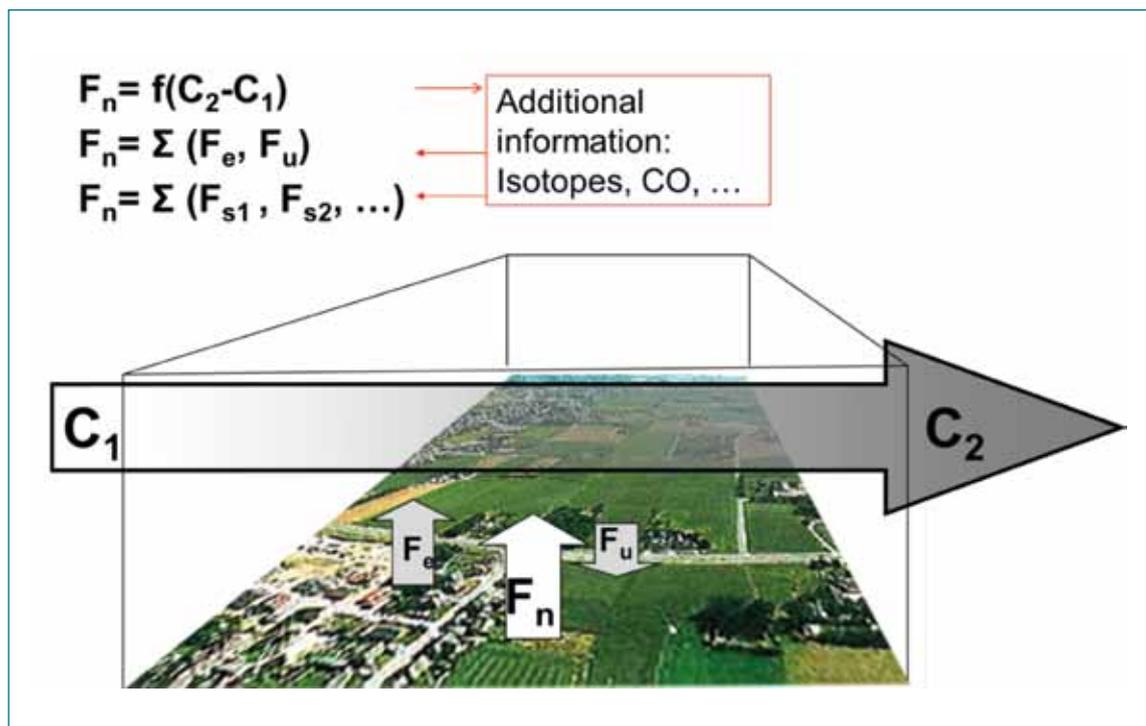


Figure 1.1.

Conceptual diagram of inverse flux estimates using atmospheric concentration measurements. The difference between the concentration of an air mass flowing into a volume ( $C_1$ ) and that of the same air mass flowing out of a volume ( $C_2$ ) is linearly related to the total net flux originating in that volume ( $F_n$ ), here assumed to come only from the surface. To deconvolve this net flux into its component fluxes (e.g. sources  $F_e$  vs sinks  $F_u$ , or various source categories  $F_{s1}, \dots$ ) requires additional information that may come from observations on isotopes and or other trace gases. Atmospheric transport models are needed to properly describe the movement of air through the volume.

The optimal set of fluxes for a region is determined by minimizing the mismatch between modelled and observed concentrations, accounting for measurement and model errors. So the method rests on three pillars: the GHG observations, the transport model combined with a priori fluxes information that simulates the concentrations at observation sites, and the inversion algorithm that adjusts the prior fluxes so as to minimize the differences between observed and simulated GHG concentrations.

Input data for inversions come from the network of GHG concentration measurement sites, until quite recently relying mostly on discrete flask sampling but now more and more also from an increasing number of in situ continuous measurement sites and from remote sensing platforms like satellites. Generally, regional fluxes derived from inverse models have smaller uncertainties upwind



of regions with denser data coverage. Also the region 'seen' by the measurements becomes larger when the observation point is located higher in the atmosphere. Uneven and sparse coverage of the network may generate additional errors in inversion results, depending on the regional meteorology. Monitoring issues will be extensively discussed in chapter two.

Transport models need to simulate wind fields and the (turbulent) mixing as close as possible to reality, but the choice of transport model does affect the inversion results. In principle, numerical weather prediction models using the extensive weather monitoring networks could fulfil this role, though they are optimised for different purposes. Arguably, more interaction between the two communities is potentially fruitful and therefore needed. A priori flux information can be derived from any of the bottom up methods discussed before, including both biogenic sources and sinks as well as anthropogenic emissions. Fossil fuel emissions are generally considered known with small uncertainties in inversions, so that their effect can be easily modelled and subtracted from the total flux to solve for regional biogenic land-atmosphere and ocean-atmosphere fluxes. While this may be a reasonable approximation at the largest scales and for the annual total flux, considerable uncertainty exists when these fluxes need to be distributed in time and space as input for the regional transport model. This can lead to strong biasing of the results, see (Gurney and al. 2005). At smaller scales uncertainties in anthropogenic emissions become comparable to those in biogenic fluxes. Finally, also inverse methodological details associated with the choice of minimization algorithm, any data filtering procedures, etc., affect the inverse results. However, in general alternative inverse estimates agree much more on the temporal variability, year-to-year anomalies or trends of fluxes than they do on the absolute flux magnitude or on the long-term mean inverted regional fluxes (IPCC 2007). For more details on the transport models, prior fluxes and inversion methodology see chapter three.

Atmospheric inversions are used in combination with estimates of surface fluxes from ecosystem models, oceanic and anthropogenic flux inventories, to quantify and improve understanding of natural carbon sources and sinks at the global scale [e.g., (Tans, Fung et al. 1990); (Rayner, Law et al. 1999); (Gurney, Law et al. 2002); (Rodenbeck, Houweling et al. 2003); (Rayner, Law et al. 2008)]. However, the uncertainties of global inversions grow quickly when zooming in to more regional scales [Baker et al., 2006]. For Europe, the estimates of biogenic land fluxes (over an area of  $1.05 \times 10^7 \text{ km}^2$ ) during 2001-2004 vary between the inversion systems of (Peylin, Rayner et al. 2005) ( $-0.3 \text{ PgC.y}^{-1}$  or  $\sim -29 \text{ gCm}^{-2} \cdot \text{y}^{-1}$ ), of (Rodenbeck, Houweling et al. 2003) ( $-0.86 \text{ PgC.y}^{-1}$  or  $\sim -82 \text{ gCm}^{-2} \cdot \text{y}^{-1}$ ) and of (Peters, Krol et al. 2010) ( $-0.12 \text{ PgC.y}^{-1}$   $\sim -11 \text{ gCm}^{-2} \cdot \text{y}^{-1}$ ). Negative and positive values correspond to  $\text{CO}_2$  uptake and sources respectively. There are even greater uncertainties at higher spatio-temporal scales because fluxes are influenced by strongly heterogeneous climatic, biogenic and anthropogenic drivers in Europe. Such issues and examples from more continental (European) and regional (Dutch national) inverse studies will be discussed in chapter 3, as the present project was one of the main drivers behind recent progress in high resolution / small domain inversions.

There is a strong need to integrate all of the three classes of methods (inventory based, flux tower based and inversion based) thus arriving at the so-called multi-constraint methodology, since none on its own can provide a complete insight in the land to atmosphere fluxes across scales. While until recently the estimates from each differed considerably, improvements in each have led to considerable convergence e.g. for Europe compare (Janssens, Freibauer et al. 2003) with (Peters, Krol et al. 2010).

### Monitoring of natural sources and sinks

Perhaps the single most important reason for being able to attribute changes in GHGs to specific areas and processes is a profound concern about the stability of natural carbon stocks. These carbon stocks form an enormous reservoir of potential CO<sub>2</sub> in the atmosphere: active terrestrial carbon stocks are estimated to amount to 2,370±125 Pg C, while more inert reservoirs (permafrost) add 1,600±300 Pg C (Ciais, Tagliabue et al. 2012). Conversion of a small fraction of current carbon stocks will amplify the anthropogenic fossil fuel caused CO<sub>2</sub> concentration rise of the last centuries, which then will potentially start a strong positive feedback on climate.

On one hand the carbon stocks are reduced directly by human activities (deforestation, peat land drainage). On the other hand, climate change itself is affecting the stability of various ecosystems that presently store large amounts of carbon: dense tropical forest may turn to savannah in drying climates (Malhi, Aragao et al. 2008) and peat lands under permafrost are thawing allowing their decomposition (Schoor, Vogel et al. 2009). Interactions between these direct human impacts – deforestation – and climate change may make the moment these globally important sinks switch from a being a sink to a source come sooner than either process alone (Nobre and de Simone Borma 2009).

Obviously, such processes need to be well presented in climate models in order to predict climate change more than several decades ahead, and monitoring is crucial for that. Also we need monitoring to allow early warning of such sink to source switches, in order to take crash mitigation actions (Shepherd, Caldeira et al. 2009) or prepare for the worst.

This motivation naturally asks for a focus of supporting monitoring systems on the natural hotspots, on the large scale ecological units that are threatened most: boreal ecosystems with much buried carbon and wet tropical forest and peat lands, the southern hemisphere oceans, etc. Regions whose significance from an earth system perspective does not coincide with their political significance. While in situ measurements of both fluxes stocks, and atmospheric GHG concentrations are slowly increasing these areas in general are still severely under sampled. The relevance of new measurements in these areas, improving our ability to constrain inversions for these regions, is high. Recent atmospheric inversions using new datasets from the region indicate that the main global terrestrial sink is located more south of the equator than previously assumed (Stephens, Gurney et al. 2007). Arctic measurements used in another inversion showed that the recent renewed increase in atmospheric growth rates of methane (after decreasing growth rates for nearly two decades) were due to anomalously high temperatures in the Arctic and more than average precipitation in the tropics, while they at the same time suggest that strong climate feedbacks from permafrost and CH<sub>4</sub> hydrates have not (yet) been activated (Dlugokencky, Bruhwiler et al. 2009).

### Monitoring for management of greenhouse gas emissions

From a policy point of view GHG monitoring is needed for the development of mitigation options and to assess their effectiveness, and for the assessment of compliance of parties to emission reduction treaties, whether national or international. These needs bring along a focus on quantifying emission trends, preferably with annual resolution. It also asks for information on source categories, i.e. emissions from specific economic sectors. More recently, there has emerged an almost natural shift in focus from national GHG policies and fitting assessments towards formulating reduction targets for the anthropogenic hotspots: large industrialized and/or urbanized areas, and supporting monitoring systems are following.

Attribution of emissions to responsible parties asks for study domains consistent with administrative boundaries. While bottom-up methods can easily conform to such sharp geographical delineations,



obviously inverse atmospheric methods cannot, due to the chaotic nature of the atmospheric dilution process. The processes involved in the mixing cannot be described exactly but have to be described by statistical means. This is also the reason why longer time-series and partially overlapping data is needed to create the redundancy needed for the statistical (inverse) methods to succeed.

The need for assessing emissions from anthropogenic hotspots as recently resulted in several attempts to quantify emissions from single cities or urban agglomerates (e.g. (Turnbull, Karion et al. 2011), (Molnar, Major et al. 2010).

### The ideal monitoring system

The long lifetime in the atmosphere of greenhouse gases of several to many years means that signals of the emissions are transported over large distances. This is an advantage for the observational system as this means that the density of observation points can be relatively sparse, but this also means that for a regional network the boundary conditions need to be taken into account as well, as sources from outside the domain and even the accumulated signal of all sources and sinks from the whole globe will have a substantial influence on the background concentration signal. This calls for a well-connected and coordinated global network consisting of a bundle of regional networks and more remote background stations in between. In this network all measurements should be of adequate precision and accuracy.

In the atmosphere the GHG signal is always a mixture of local, regional and global sources and sinks, that can be natural and man-made. This has as a consequence that it is not possible to concentrate on just the atmospheric signal of anthropogenic influences but that also the (semi-) natural emissions with all the inherent complexity connected to it have to be taken into account in the (inverse) modelling and prior and posterior fluxes.

In many cases measurements of the isotopic ratios of the atmospheric GHGs can help in discriminating between different source types; these observations are however at this moment quite sparse and expensive. Further developments in instrumentation (and modelling of isotopic composition and source characterisation) is needed here.

Another important design parameter for the global network besides binding the regional networks focused at anthropogenic hotspots should be the (early) detection of natural emission changes due to climate change. This requires an extension of the current network and focus of satellite missions on currently under-sampled regions like the tropics and the (sub)arctic and permafrost regions of the world.

Attribution of changes in greenhouse gas levels to specific areas and processes is an important prerequisite to monitor treaty compliance, policy effectiveness, and the stability of natural greenhouse gas sinks. Nevertheless, and despite repeated pleas (Tans, Bawkin et al. 1996) and international formal agreements (UNFCCC 2004), it is very hard to fund long term, uninterrupted monitoring of GHGs (and other climate variables) that helps us understand how the earth system changes.

Monitoring programmes are being designed at complementary scales ranging from global (e.g. Geo Carbon, (Ciais, Dolman et al. 2010) to continental (e.g. the ICOS initiative in Europe) to national (the various ICOS national contributions). Most of these are still research oriented and funding is project based resulting in poor guarantees in terms of long term continuity. Instead the need is emerging

to move to more operational modus operandi, in which more service oriented agencies take over responsibilities for monitoring networks and associated data archiving and dissemination activities. First moves in this direction are apparent with meteorological services taking responsibilities for part of the tasks (like DWD in Germany). Even private weather services apparently see commercial potential in gathering GHG measurement data and building information services around these (e.g. EarthNetwork).

### Only the atmosphere matters

From a climate change perspective only the *atmospheric* concentrations fraction of GHGs matters in the end, or when we limit ourselves to carbon only the size of the *atmospheric* carbon pool matters. Carbon dioxide is responsible for the largest fraction of man-made positive climate forcing. More than 90% of the annual increase in radiative forcing from GHGs since 2000 is due to CO<sub>2</sub> alone. Evidence from carbon isotopes (<sup>14</sup>C depletion) indicates that more than 75% of the increase in CO<sub>2</sub> over the past century can be attributed to fossil fuel burning. Mitigation of these emissions is costly, the more so the faster and stronger the reductions are required, and may require a profound change of our life styles. Therefore there will be a strong tendency to cling to the more favourable side of the uncertainty range of flux estimates, natural or anthropogenic.

In the assessment of the strength and future evolution of natural carbon sinks (in forest, whether tropical, temperate or boreal) the high end estimates will be more readily accepted in society than the low end ones, as they relax the targets we need to set for fossil fuel emission reduction. Emission trading schemes, widely accepted as one of the more practical and fair policy instruments, explicitly monetize emissions and reductions thereof. Again, wherever there is room to do so, room created by the imperfection of emission estimates, reduction claims will more readily be overestimated than underestimated, simply because it saves money to do so.

Since the atmosphere is the only place where carbon dioxide or other GHGs matter, it is *the* place to measure stocks, i.e. concentrations. From *these* concentrations changes we need to deduce fluxes and attribute these to specific sources or sinks, to specific regions, to specific actions we take. We need this ultimate check on any other flux estimate we make. Discrepancies between bottom up and top down estimates must be reduced as much as possible for us to have any confidence in our projections of radiative forcing and thus in our projection of climate change.



## 2. The monitoring system for greenhouse gas emission verification

### The current network vs the ideal network

The ideal network for greenhouse gases emission verification would consist of a cost-effective number of stations at optimum positions to derive the flux information at the highest desired precision and resolution in space and time. The current situation of the network is unfortunately quite far from this.

The problem in designing and implementing a more optimal network is that one needs to first state realistic requirements for resolution and precision, that logistic limitations are always imminent and that the methodology using atmospheric dilution processes will always be limited in its maximum precision due to inherent uncertainties that are not known at design time. The attempts of network design thus far using advanced inverse modelling techniques come to answers that depend more on the model uncertainty and structure than on the actual network structure itself (refs). Synthetic experiments deploying perfect models and observations are not realistic and lead to the conclusion that a network consisting of only one observation point would be enough to get perfect emission estimates for a very large region. One has to take the uncertainties in both model and observations into account; the problem with uncertainties is that they are unknown and that we have to make assumptions on their size and structure. This calls for an recursive network design where we build experience through step by step increasing the complexity and density of the network and inverse models in order to evaluate the precision and resolution of the fluxes that can be derived from the network using uncertainty estimates and model errors based on the observations.

Until now the network in Europe has been built up from a myriad of national and European projects and stations are run by a large biodiversity of institutes. The next step in the ground based greenhouse gas observation network for Europe will be the ICOS network (<http://www.icos-infrastructure.eu>), where a more dense network of coordinated observations will be set up. ICOS is foreseen to become operational in 2012.

In the observation strategy we can choose from several observational techniques, that all have their own specific characteristics and advantages.

### In situ monitoring of greenhouse gas concentrations

#### Ground based observations

For fixed ground based observations one can choose between continuous and discontinuous sampling in time. Discontinuous sampling allows for off-line analysis at a central location, thereby minimizing the amount of expensive and sensitive equipment. The low-cost and simple collection of e.g. flask or bag samples allows sampling even under primitive conditions and at remote locations without facilities as electricity and network. Disadvantages can be that samples change in composition during transport to the analysis facility and the relatively low frequency of the observations. Also care should be taken to avoid biases through the sampling strategy, e.g. by taking samples at the wrong time or location.

(Semi-)Continuous sampling requires equipment at each sample location. Main advantage is the higher frequency with which observations are obtained. Clear disadvantages are the higher overall cost and that the sampling location needs to offer more facilities like shelter, electricity,

data connection and sometimes climate control. If the measurements conditions are chosen and maintained well, the precision and accuracy of the measurements can be comparable to laboratory standard.

For greenhouse gas observations the WMO issued a set of recommended target accuracy and precision for the most important greenhouse gases. The current recommendations are listed in the following table.

Table 2.1.

Recommended inter-laboratory network comparability according to WMO GAW (GAW Report No 186)

Trace gas component	Approximate present average global atmospheric concentration (and at Cabauw <sup>1</sup> )	Recommended inter-laboratory comparability
CO <sub>2</sub>	388 ppm	± 0.1 ppm (±0.05ppm southern hemisphere)
CO	50-300 ppb <sup>2</sup> (117ppb)	± 2 ppb
Δ <sup>14</sup> C – CO <sub>2</sub>	100 ‰	± 1 ‰
CH <sub>4</sub>	1745 ppb (1983 ppb)	± 2 ppb
N <sub>2</sub> O	314 ppb (323 ppb)	± 0.1 ppb
SF <sub>6</sub>	7.1 ppt (6.8 ppt)	± 0.02 ppt

1) winter 2008 (Vermeulen, Hensen et al. 2011)

2) CO concentration are spatially highly variable, ranging from virtually zero in pristine conditions (S-Hemisphere) to over 300ppb in heavily industrialised areas

In general point observations have limited representativity and may be influenced by (local) sources or flow patterns that are and/or cannot be resolved by current transport models. The choice of location of the observation point is therefore important, so that the measurement is taking place in an environment that is representative of the source area that it has to collect data from or that the local influence can be characterized or minimized. Minimization of this influence can be obtained by using stations placed on top of hills or by using tall towers. The use of tall towers has the added advantage that one can sample at several heights in order to derive vertical gradients so that the average (bulk) mixed layer concentration can be measured. In the Netherlands two tall towers are operational: Cabauw (Vermeulen, Hensen et al. 2011), see below) and Lutjewad (van der Laan, Neubert et al. 2009). Mountain or hills stations have some disadvantages in that their height is difficult to represent in transport models and that they can be periodically influenced by upslope or downslope winds.

#### Vertical profile observations

Vertical profile observations of GHG concentrations can take place using the same remote sensing techniques that are deployed in satellites using optical techniques. There are active methods deploying light sources, usually lasers systems, like LIDAR and DIAL that probe the lower to middle troposphere. The accuracy and precision of these methods for GHG concentration profiling are quite limited, but if they are connected to co-located observations using more precise techniques this problem can be corrected for. The presence of water vapour in the air is a limiting factor for the precision of these observations, as well as presence of clouds.



Passive techniques like FTIR can be used by solar tracking observations. In this way the whole vertical column between sensor and the sun is probed. These observations are just like most satellite observations limited to cloud-free conditions and periods where the sun is at higher zenith angles.

#### Mobile (airborne) observations

Airborne observation allow to sample the atmosphere in four dimensions, which in principle allows to obtain a more representative picture of the distribution of the concentration. Depending on the size of the aircraft the equipment can be anything between continuous, multicomponent and very accurate, and basic grab sampling using automated or manual flask samplers for off-line analysis, or a combination of these techniques. Disadvantage of the airborne sampling are the high costs of operations and the fact that sampling is limited to specific flight conditions and by air traffic restrictions. The cost of operations can be reduced by using unmanned aircrafts or balloons, but these platforms are very restricted in the size of the equipment that can be carried.

The representations of aircraft observations in transport models is very challenging, but is important to improve especially the vertical transport mechanisms in those models.

#### Remotely sensed greenhouse gas concentrations

Accurate detection of long-lived greenhouse gases from space is highly challenging, because of the small concentration gradients of these gases and consequently high requirements on the accuracy and precision of the measurements. The SCIAMACHY instrument has been measuring CO<sub>2</sub> and CH<sub>4</sub>, among other gases, since the year 2002. CO<sub>2</sub> is not an official data product of SCIAMACHY, but could be retrieved from measurements that were initially meant for cloud detection. Much progress has been gained on the retrieval of CO<sub>2</sub> from SCIAMACHY over the years. Nevertheless it has not reached the accuracy level needed for global source and sink estimation. The most important remaining hurdle is to account for cirrus and aerosol scattering along the light path (Houweling, Hartmann et al.), (Butz, Hasekamp et al. 2009).

In the case of CH<sub>4</sub> the situation is much more favourable thanks to the so-called proxy approach (Frankenberg, Meirink et al. 2006), in which CH<sub>4</sub> is determined from the observed ratio of CH<sub>4</sub> and CO<sub>2</sub>. The advantage of this approach is that errors due to aerosol and cirrus scattering cancel out to a large extend. Initially the CH<sub>4</sub> measurements from SCIAMACHY pointed to significantly underestimated CH<sub>4</sub> emissions from tropical forests (Frankenberg, Meirink et al. 2005). Later it was found that a substantial fraction of the increased CH<sub>4</sub> concentrations was explained by inaccuracies in the CH<sub>4</sub> and H<sub>2</sub>O spectroscopy in the 1.6 micron wave length region (Frankenberg, Bergamaschi et al. 2008). Current inverse modelling estimates on the basis of SCIAMACHY CH<sub>4</sub> show residual enhancements of tropical CH<sub>4</sub> emissions, within a range that can be explained by difficulties to account for methane emissions from tropical wetlands and wet soils (Bergamaschi, Frankenberg et al. 2009).

Since February 2009 the Japanese Greenhouse Gas Observing SATellite (GOSAT) is measuring CH<sub>4</sub> and CO<sub>2</sub>. GOSAT is the first satellite mission that is specifically designed to measure long-lived greenhouse gases. In comparison with SCIAMACHY, GOSAT has a much higher spectral resolution, which allows a combined retrieval of greenhouse gases, aerosol and cirrus. Current attempts to obtain high quality retrievals of CO<sub>2</sub> and CH<sub>4</sub> from GOSAT highlight the difficulty to measure those compounds at sufficient accuracy. Ground-based Fourier transform spectroscopy (FTS) assists in the verification of GOSAT measurements and points to remaining offsets and scatter in the data, which gradually improve thanks to the on-going research efforts of several groups involved.

### Observations at Cabauw tall tower.

Cabauw tower (4.927° E, 51.971° N, -0.7ma.s.l.) is a steel structure that rises up to 213ma.g.l. It is located in the centre of The Netherlands about 25 km southwest of the city of Utrecht and is operated by KNMI. In 1992 the Cabauw tower was equipped by ECN for greenhouse gas observations (Vermeulen, Hensen et al. 2011). Cabauw tall tower has been used since then in different gradually improved equipment configurations. In November 2004 a new set of equipment has been placed at Cabauw in the framework of the CcSP-ME2 and CHIOTTO (EU 5th framework programme) projects. The old GC system was replaced by an Agilent 6890N GC with FID and ECD detectors for CH<sub>4</sub>, CO, N<sub>2</sub>O and SF<sub>6</sub> measurements. The CO<sub>2</sub> analyser Siemens Ultramat NDIR was replaced by a Licor 7000 NDIR analyser.

In 2006 the Cabauw equipment was supplemented with a <sup>222</sup>Rn monitor from ANSTO (Zahorowski et al., 2004), sampling from 200m, followed by a second <sup>222</sup>Rn monitor of the same type in 2007, sampling from 20m.

The observations at Cabauw show a complex pattern caused by the influence of sources and sinks from a large area around the tower with significant contributions of sources and sinks at distances up to 500–700 km, figure 2.1. The concentration footprint area of Cabauw is one the most intensive and complex source areas of greenhouse gases in the world. Despite this, annual mean trends for the most important greenhouse gases, compatible with the global values derived using the global network, can be reproduced from the measured concentrations at Cabauw over the entire measurement period, with a measured increase in the period 2000–2009 for CO<sub>2</sub> of  $1.90 \pm 0.1$  ppmyr<sup>-1</sup>, see figure 2.2, for CH<sub>4</sub> of  $4.4 \pm 0.6$  ppb yr<sup>-1</sup>, for N<sub>2</sub>O of  $0.86 \pm 0.04$  ppb yr<sup>-1</sup>, and for SF<sub>6</sub> of  $0.27 \pm 0.01$  ppt yr<sup>-1</sup>; for CO no significant trend could be detected.

The influences of strong local sources and sinks are reflected in the amplitude of the mean seasonal cycles observed at Cabauw, that are larger than the mean Northern Hemisphere average; Cabauw mean seasonal amplitude for CO<sub>2</sub> is 25–30 ppm (higher value for lower sampling levels). The observed CH<sub>4</sub> seasonal amplitude is 50–110 ppb. All gases except N<sub>2</sub>O show highest concentrations in winter and lower concentrations in summer, N<sub>2</sub>O observations show two additional concentration maxima in early summer and in autumn.

Seasonal cycles of the day-time mean concentrations show that surface concentrations or high elevation concentrations alone do not give a representative value for the boundary layer concentrations, especially in winter time, but that the vertical profile data along the mast can be used to construct a useful boundary layer mean value. The variability at Cabauw in the atmospheric concentrations of CO<sub>2</sub> on time scales of minutes to hours is several ppm and is much larger than the precision of the measurements (0.1 ppm), see figure 2.3.

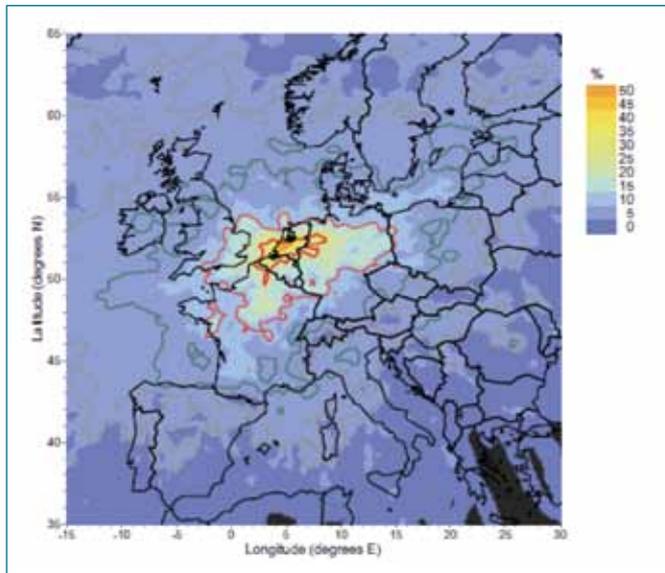


Figure 2.1.

Total hourly concentration footprint (2008) for Cabauw 200m sampling level. First thick red contour contains the area with 25%, next thin red 50%, next thick grey 75%, next thin grey 95% of total potential footprint. Colour scale is percentage of potential footprint per pixel relative to the maximum pixel value

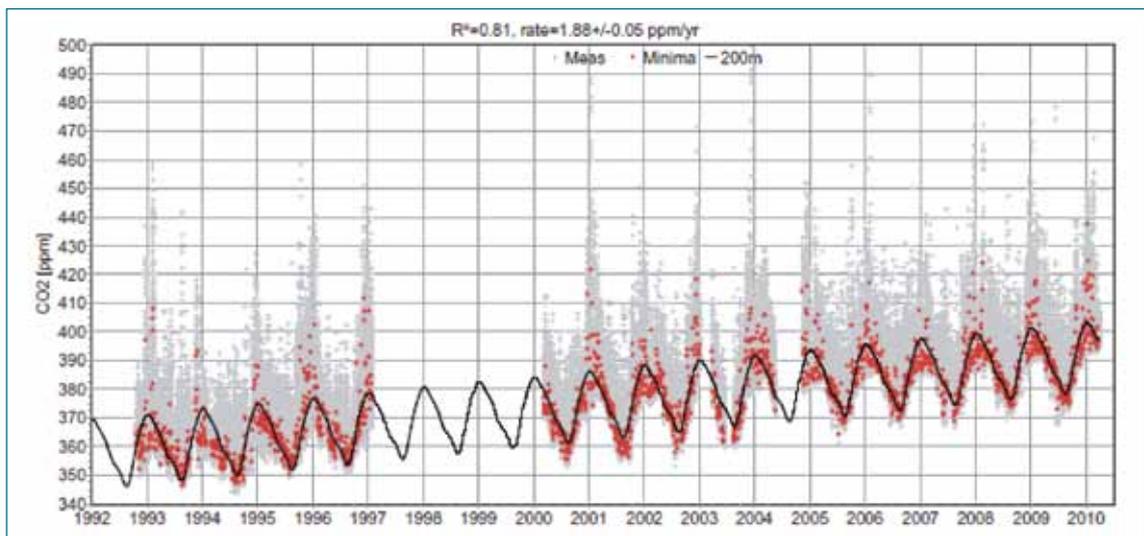


Figure 2.2.

Time series (1992-2010) of  $\text{CO}_2$  at 200m: all values (dark grey crosses), daily 50%-trimmed mean values (coloured dots), fitted harmonic function (black line, method B) and confidence interval for the harmonic fit, computed from the standard deviation of the daily trimmed values (light grey area)

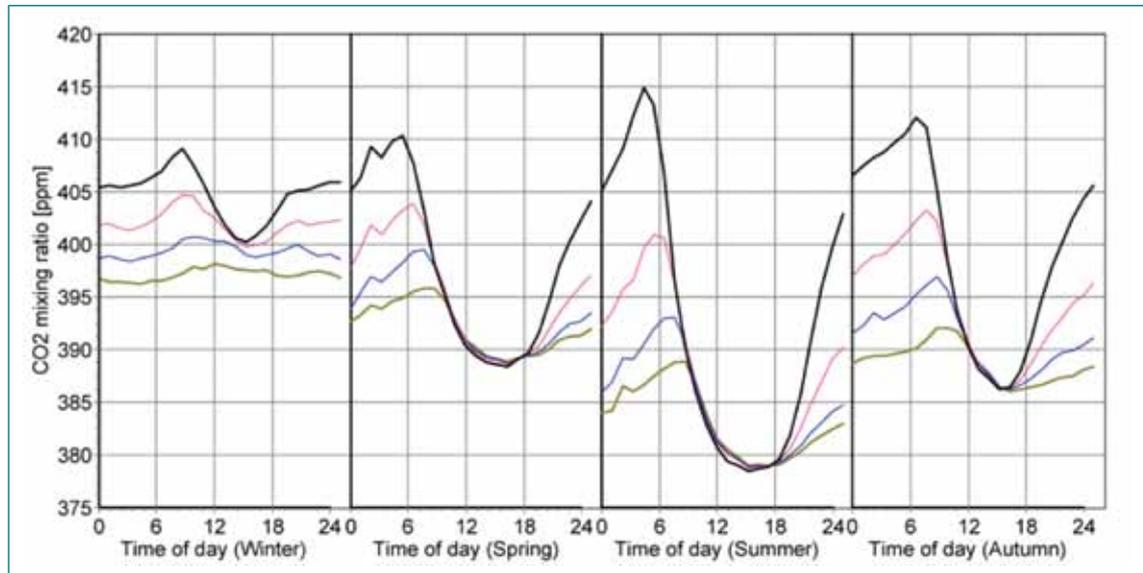


Figure 2.3.

Average diurnal vertical concentration profiles per season at Cabauw for the period 2005–2008 for CO<sub>2</sub>. Seasons are defined as Winter=Dec-Feb; Spring=Mar-May; Summer=Jun-Aug; Autumn=Sep-Nov

## Direct observations of regional fluxes

Several techniques exist to directly determine fluxes at larger scales and the objective of these can be two fold. One, to retrieve estimates of the sensible heat fluxes at scales relevant for boundary layer development (which directly affect GHG concentrations) these can be measured either through eddy covariance measurements at elevations well above the surface layer or through large aperture scintillometry with path lengths of several kilometres. Alternatively, GHG fluxes can also be measured directly at regional scales of 10-10<sup>2</sup> kms from low flying aircraft a method currently feasible for CO<sub>2</sub> and under development also for CH<sub>4</sub>.

### Turbulent flux observations at the 200 m meteorological tower at Cabauw

At many sites over the world CO<sub>2</sub> surface fluxes are estimated at eco-system level by performing turbulence observations at a few meters above canopy height. In this project we are interested in flux estimates representative for the regional scale. By installing turbulence instruments at several levels in the 200 m meteorological tower at Cabauw we are in a unique position to perform long-term flux on this regional scale. In the course of the project existing equipment was modernized, the number of instrumented levels was increased and instrumentation was harmonized. Crucial additional observations of CO<sub>2</sub> concentration profiles were performed by ECN.

Fundamental studies on the interpretation of the observations have been performed by (Schuitmaker 2009) on the behaviour of the temperature observed with a sonic anemometer/thermometer and by (Schalkwijk, Bosveld et al. 2010) on the correction for low frequency flux loss in elevated turbulent flux observations. Interpretative studies with the tower flux observations have been performed by (Casso-Torralba, Vilà-Guerau de Arellano et al. 2008) on atmospheric budgets for three specific days and by (Werner, Bosveld et al.) looking at longer time series showing that reliable regional surface flux estimates of CO<sub>2</sub> can be made when horizontal advection can be ignored. For sensible and latent heat flux these advection terms can be reasonably estimated from numerical weather prediction/assimilation systems which have been improved over the years. For CO<sub>2</sub> this still remains to be shown.



### Scintillometer observations of regional fluxes

The dynamics of the boundary layer play an important role in the link between GHG emissions and the ensuing concentrations. The most important variable in this respect is the boundary-layer height, which is largely determined by the surface sensible heat flux. In order to study the variability of this main driver, a new operational network of optical scintillometers over different types of terrain (providing sensible heat fluxes) has been setup. The surface types covered forest pasture, fen meadow, mixed agriculture and city. Scintillometers have been used because they provide fluxes at a scale (order of kilometres) that is compatible with the scale of boundary-layer processes.

Apart from the operational measurements, that have been carried out from 2006 until 2010, background research has been performed on the interpretation of scintillometer signals (and turbulent signals in general) over heterogeneous terrain (Moene and Gioli 2008; Moene and Schuttemeyer 2008) Furthermore, the data have been used in model validation studies (Steenefeld, Hartogensis et al. 2008; Steenefeld, Tolk et al. 2011), see relevant section of this report.

At the Cabauw Experimental Site for Atmospheric Research (CESAR) we are in a position to measure with a scintillometer over a very long path due to the presence of two tall towers within a distance of 10 km. In the period 2002-2003 an eXtra Large Aperture Scintillometer (XLAS) (built by WUR and KNMI) was operated over a path of 10 km at a height of 40 m between the 200 m meteorological tower of Cabauw and the TV tower of IJsselstein (The Netherlands). Analyses showed that during dry periods in summer sensible heat fluxes were large enough to cause significant saturation of the scintillometer signal. From 2008 onward an XLAS (Kipp&Zn) is operated over the same path but at a height of 60 m to decrease the problem of saturation. At the same time turbulent observations are performed at several levels along the meteorological tower among which the 60 m level. (Braam 2008; Braam, Bosveld et al. 2011 (subm) studied the relation between the 60 m XLAS derived sensible heat flux and the surface sensible heat flux and found good agreement when estimating the regional scale surface sensible heat flux from the sum of eddy-covariance observation at 60 m height and the rate of change of heat content in the atmospheric column below 60 m.

### Monitoring fluxes of heat and CO<sub>2</sub> from small aircraft

Aircraft measurements of turbulent fluxes are generally being made with the objective to obtain an estimate of regional exchanges between land surface and atmosphere, to investigate the spatial variability of these fluxes, but also to learn something about the fluxes from some or all of the land cover types that make up the landscape. Here we'll briefly discuss the primary technicalities of obtaining fluxes, and subsequently focus on various approaches to address such issues for especially CO<sub>2</sub> fluxes.

In comparison with tower based eddy correlation monitoring, measuring turbulent fluxes from small aircraft flying at low altitudes is complicated by two issues. First, the airframe itself distorts the flow at the turbulence sensor, second the aircraft motions are affected by the turbulence itself. Furthermore, both effects interact dynamically. Aircraft motions must and are monitored at high frequencies using some IGPS system, such that in principle the aircraft velocity vector can be subtracted dynamically from the turbulent wind vector. Semi empirical relations exist to estimate the flow distortion near the turbulence sensor allowing correction of especially the upwash induced by the wings. Though various approaches to both correction procedures existed in grey literature, (Vellinga, Dobosy et al. subm) propose and implement a consistent set of algorithms for post processing of airborne turbulence data, as well as for estimation of the empirical parameters needed in that. They clearly demonstrate a very significant improvement of the raw wind field measurements as well its effect on the eventually derived flux magnitudes.

The result are airborne observed surface fluxes that are very well comparable to tower based estimates (Gioli, Miglietta et al. 2004; Vellinga, Gioli et al. 2010) when obtained at low altitudes. When flying above the surface layer flux divergence may become significant (Vila –Gerau et al., 2005) and may hinder interpretation in terms of surface fluxes. However, such measurements can still be very valuable for direct comparison to mixed layer fluxes as computed in 3D atmospheric transport models (e.g. (Sarrat, Noilhan et al. 2007; Sarrat, Noilhan et al. 2009).

To assess regional fluxes from aircraft observations and to scale these to larger areas various techniques have been developed. Aircraft measurements of regional net ecosystem exchange (NEE) can be spatially extrapolated using a combination of flux measurements made by aircraft, half-hourly eddy covariance data from towers, half-hourly weather data and detailed information on regional land use in a simple spatially explicit model (e.g. (Miglietta, Gioli et al. 2007). Similar scaling approaches using satellite measurements have been developed for heat fluxes (Miglietta, Gioli et al. 2009). Alternatively, the landscape is first stratified into more or less homogeneous areas over which aircraft fluxes are averaged and then these can be scaled to the region when the fraction of these landscape elements in the region is known (Vellinga, Gioli et al. 2010).

To address spatial variability of fluxes more directly direct mapping approaches have been successfully developed by e.g. (Mauder, Desjardins et al. 2008), allowing almost grid based mapping of surface fluxes. Vellinga et al. (in prep) developed a monitoring and analysis strategy allowing characterization of both spatial variability and seasonal dynamics of regional surface fluxes in the Netherlands, creating a dataset that is unique in its spatial and temporal coverage.

Finally, many groups are or have been working on estimation of fluxes for specific homogeneous land cover types in a region. Whereas some use conceptually questionable approaches for disaggregation of blended signals, based on using footprints assigned to very short turbulence fragments (Kirby, Dobosy et al. 2008), others use more sound approaches but failing to present sound proofs of concept (Ogunjemiyo, Kaharabata et al. 2003). (Hutjes, Vellinga et al. 2010) developed a method that relies on using a footprint model to determine which part of the landscape the airborne flux observation refers to, using a high resolution land cover map to determine the fractional covers of the various land cover classes within that footprint, and finally using multiple linear regression on many such flux/fractional cover data records to estimate the component fluxes. A systematic proof of concept for both linear aggregation of component fluxes into a regional average as well as for disaggregation of regionally blended flux signals is presented in conjunction with a case study showing the value of the latter.

The usefulness and added value of airborne flux observations has been demonstrated in several integrative fluxes addressing carbon budgets at regional scales, both through data based analysis in direct relation to the actual land cover maps (Dolman, Noilhan et al. 2006), (Dolman, Gerbig et al. 2009), as well in validation of atmospheric transport characteristics in 3D high resolution coupled land-atmosphere model (e.g. (Sarrat, Noilhan et al. 2009), (Sarrat, Noilhan et al. 2009), (Lauvaux, Gioli et al. 2009).



## Monitoring of Atmospheric Boundary Layer dynamics

Boundary layer properties affect the vertical and horizontal transport of greenhouse gases. Direct measurement of vertical turbulent mixing characteristics are relevant to the improvement of parameterisations of these, needed in atmospheric transport models. Monitoring of boundary layer heights is highly relevant for the interpretation of GHG concentrations as its growth or shrinkage dilutes/concentrates any signal from surface fluxes.

### Determination of the mixing layer height by a ceilometer

The mixing layer height (MLH) determines the volume in which heat, momentum, atmospheric gases and aerosols are transported from and to the surface due to mixing. In general the aerosol concentration in the boundary layer is higher than in the relative clean free troposphere aloft. The presence of aerosols in the atmosphere can be detected in the backscatter profiles of LIDAR systems. Therefore, a (strong) negative gradient in the measured backscatter profile of a LIDAR generally marks the top of the boundary layer. An algorithm for the routine determination of MLH from the commercial Vaisala LD-40 ceilometer (a low power solid state LIDAR) was developed at KNMI and evaluated within the BSIK ME2 project (de Haij, Wauben et al. 2006), (Wauben, Klein Baltink et al. 2006), (Wauben, de Haij et al. 2008).

The LD40 ceilometer is deployed at more than 30 stations in KNMI's synoptical network. However due to the network design the backscatter profiles were stored only for stations Cabauw and De Bilt. For the ME2 project five additional stations were selected for which backscatter profiles were stored locally. This configuration is presented in figure 1, along with measured backscatter data. The LD-40 is a bi-axial system, hence the beams of the transmitter and receiver do not overlap completely in the lowest range, up to 60 m the overlap is even zero. Therefore detection of the MLH is only feasible from 90 m agl. onwards. Backscatter profiles are reported by the LD-40 every 15 seconds, together with the lowest three cloud bases.

To retrieve the MLH from the backscatter profiles a wavelet algorithm is applied to the 10 minute averaged profile within a vertical range of 90 to 3000 m. For each averaged profile the top of two significant aerosol layers can be detected. This gives the opportunity to detect the mixing layer height as well as the top of a secondary aerosol layer, like e.g. the residual layer. The wavelet MLH method uses a minimum step size of 15 m. i.e. 2 range gates of the backscatter profile. The top of the first layer is detected at the first range gate at which the scale averaged power spectrum shows a local maximum which exceeds a threshold value. This value is empirically chosen based on the analysis of several cases with both well pronounced and less clearly pronounced mixing layer tops.

The jump in backscatter, centered on the MLH level is used as a measure for the quality of the detection. The threshold values for the different quality index classes are to some extent chosen arbitrarily, and are estimated by assessing a number of different cases. A disadvantage of this quality index is that it is independent of the absolute value of the backscatter. A statistical analysis of the occurrence of detection and the associated quality is presented in figure 2.

The wavelet MLH detection results were compared to other methods for the detection of the MLH, e.g. from radiosonde ascents in De Bilt, and wind profiler and uv-lidar MLH estimates in Cabauw. From the intercomparison and statistical analysis we conclude that MLH detection is feasible by the LD40 but a number of limitations exist. The main findings can be summarized as follows:

1. The reliability of the MLH detection is strongly connected to the variability of the aerosol backscatter signal in the vertical profile in the mixing layer. Profiles that show a fairly constant and sufficient amount of aerosol backscatter result most of the time in a reliable detection of MLH. This mainly occurs when the mixing layer grows not too deep, e.g. in a shallow wintertime mixing layer. Especially in strong convective conditions in spring and summer, the vertical range of MLH detection by the LD-40 is limited.
2. The afternoon decay of the convective mixing layer is a problem for LD-40 MLH estimation. The large amount of aerosol in the residual layer on top of the decaying mixing layer lowers the contrast with the mixing layer and hence decreases the possibility of a correct MLH detection.
3. The algorithm has a problem with the detection of a very shallow MLH. This is especially observed during periods with a nocturnal (stable) layer. The problem is probably caused by the lowest detection height of the LD-40 (i.e. 90 m) and perhaps also by the poor numerical resolution and truncation of the backscatter data in the KNMI LD40 file format.

KNMI will upgrade its ceilometer network in the coming years. The (automated) detection of the MLH is one of the requirements that will be taken into consideration.

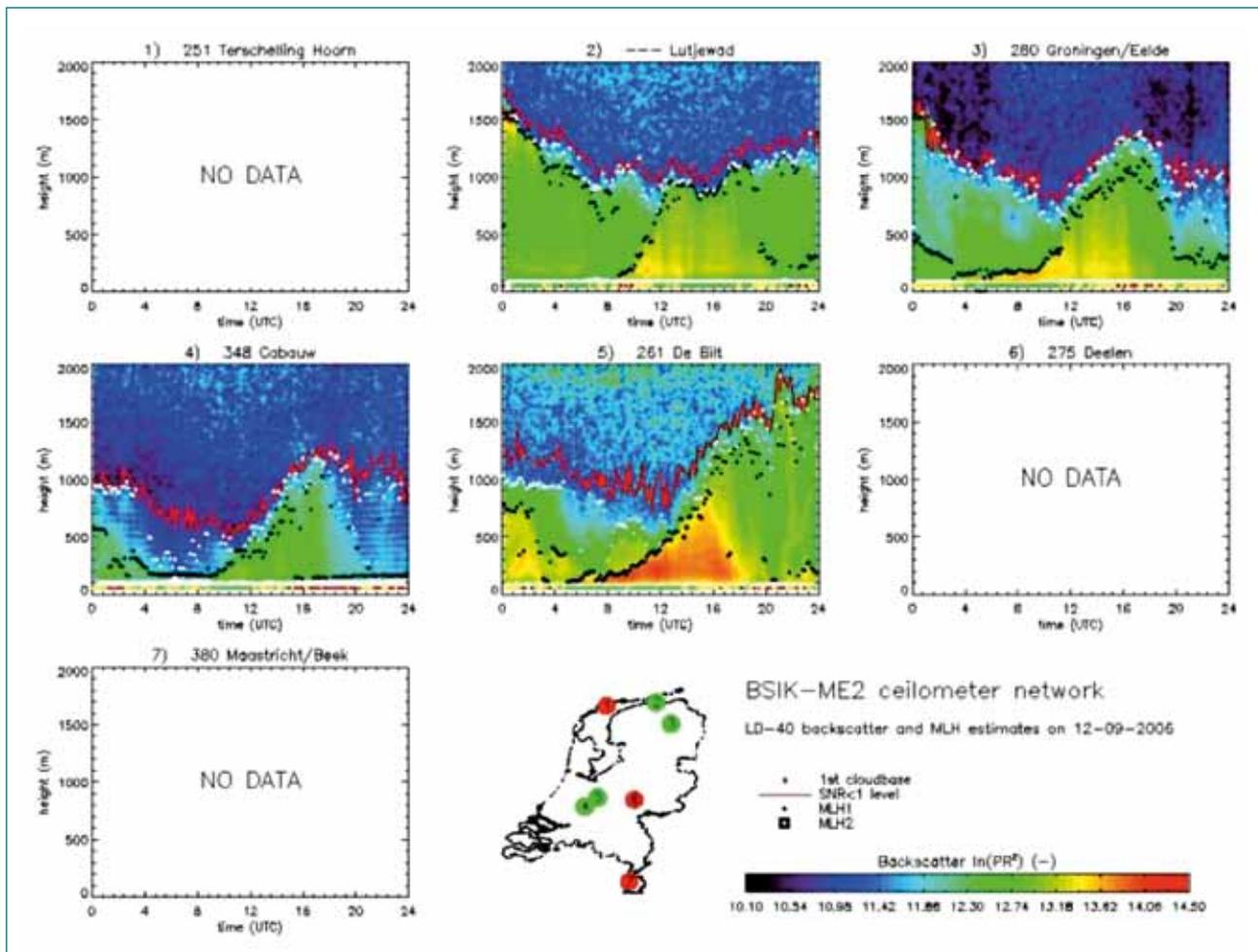


Figure 2.4.

The ME2 ceilometer network stations (lower panel), backscatter time-height plots and the detected MLH (black dots). The typical diurnal cycle in the MLH is clearly visible.

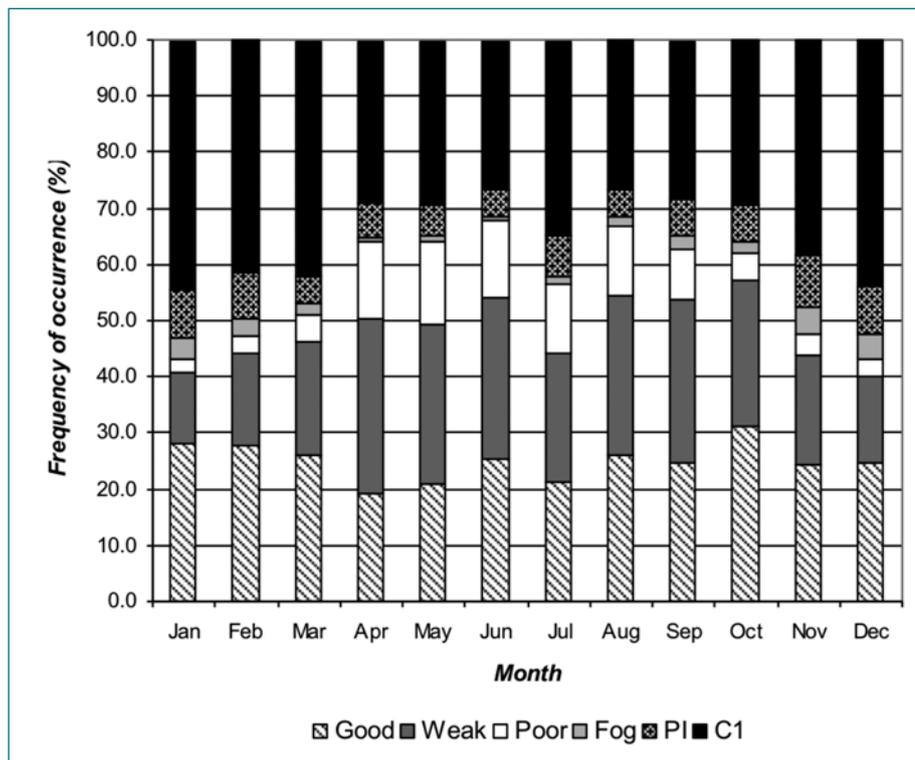


Figure 2.5.

Monthly mean detection rates for the wavelet MLH in De Bilt, period 2000-2005. Successful MLH detections are subdivided in the three quality classes: 'Good', 'Weak' and 'Poor'. Failing determination of the Wavelet algorithm ascribed to either 'Fog', 'PI' (precipitation) and 'C1' (clouds) are shown as well.

#### High-resolution boundary layer dynamics by Raman lidar

The Raman lidar technique has become the current standard for quantitative aerosol profiling over the past decade as it is a robust technique able to provide backscatter and extinction profiles without critical assumptions (Ansmann, Wandinger et al. 1992). At the same time, the Raman lidar technique offers the possibility to use the same emitter and receiver set-up to measure other atmospheric parameters by addition of detection channels for non-elastically Raman scattered light by specific molecules such as water vapour. In this way the water vapour mixing ratio can be measured. Moreover, recent developments have shown that aerosol microphysical parameters, such as effective particle size and complex refractive index, can be estimated from a minimum set of optical property profiles consisting of backscatter profiles at least three wavelengths and extinction profiles at two wavelengths profiles based (Muller, Wandinger et al. 1999; Muller, Wandinger et al. 1999; Veselovskii, Kolgotin et al. 2005) For detection of cloud phase and aerosol particle non-sphericity, the particle depolarisation ratio is an indispensable parameter that can be measured also within the same instrument since it uses a (linearly) polarised laser source.

Caeli, the CESAR Water Vapour, Aerosol and Cloud lidar, was set up as a multi-wavelength Raman lidar (Apituley, Wilson et al. 2009). For monitoring purposes, Caeli was designed as a highly-autonomous system, that can be kept running 24/7 after it has been started up by an operator. However, since it will in general not be very useful to keep the system running under persistent 'bad weather' conditions (i.e. extended periods of fog, low clouds, precipitation), the system can be shut down remotely when the atmospheric conditions become unfavourable for measurements, or automatically if a technical problem arises.

The high power of the system, realised by a high-power laser and large aperture receiving telescopes, enable Caeli to measure boundary layer dynamics at high resolution. Data is acquired at 10 second time resolution and 7.5 meter vertical resolution. An example of high-resolution water vapour measurements is shown in Figure 2.6.

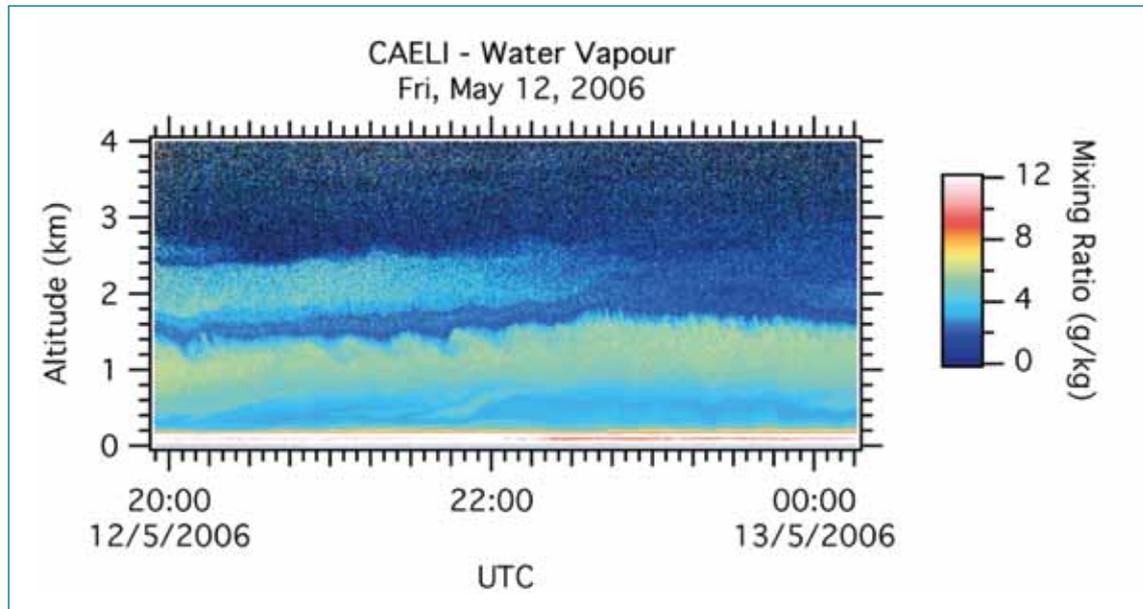


Figure 2.6.

Example of high-resolution water vapour profiles measured by Caeli, the CESAR Water Vapour, Aerosol and Cloud Lidar. The time vs. height plot shows the water vapour to dry air mixing ratio at 10 second time and 7.5 meter height resolution. A moist layer that disappears over time can be seen above the moist boundary layer. A wave appears in the boundary layer just after 21:00 UTC.

#### Boundary layer monitoring with a Doppler polarimetric radar

Identifying trace gases and aerosols and estimating their concentration is an important issue. Further, it is necessary to retrieve the dynamics of the boundary layer to quantify their displacement. Consequently, time series of wind profiles are valuable measurements. They can be performed with a Doppler sensor: lidar or radar. The lidar can measure the Doppler velocity of aerosols and cloud droplets. The radar can measure the Doppler velocity of raindrops, ice crystals and refractivity index variations due to turbulence and thermals. Emphasis was given to the study of radar wind retrievals. At Cabauw, two radars, capable of measuring wind profiles, are the operational wind profiler of KNMI and the TU-Delft research radar, TARA.

The wind profiler provides hourly-averaged profiles in routine mode with a height resolution of 100 m. Since TARA has three beams to profile the troposphere, investigation was made to increase the accuracy and the resolution of radar wind retrievals. Signal processing techniques have been substantially improved (Unal 2009), (Unal 2008), (Unal 2006). In particular, the use of polarized signals allows the separation of atmospheric echoes from non-atmospheric echoes. Hence, the unwanted signals, which are numerous in the boundary layer, can be suppressed. With the new processing, the sensitivity of the radar has been increased, which is valuable to detect the weak echoes of the refractivity index variations. With the FM-CW technology, the height resolution is a variable. It was chosen to be 7.2 m for boundary layer measurements. When thermals and downdrafts occurred, the achieved time resolution of the vertical wind is 18 s. With this high space-time resolution, the estimate of the accuracy of the vertical wind in clear air conditions varies from 0.6 to 0.8 m s<sup>-1</sup>. A mean deviation of 0.3 m s<sup>-1</sup> on the 24 min-averaged vertical wind was found



comparing all the wind sensors present in Cabauw during the IMPACT campaign (Unal, Arabas et al. 2009). This comparison was done in the context of the activation process (transformation of atmospheric aerosol particles into cloud droplets) where detailed knowledge of the spatial and temporal variability of the vertical wind (especially close to the cloud base) is needed. Concerning the horizontal wind, the time resolution of 10 min is selected to calculate the mean horizontal wind nearly free of turbulence effect and bias due to precipitation. There is a good agreement on the 10 min-averaged horizontal wind between the radar and rawinsondes.

### 3. An inverse modelling system for greenhouse gas emission verification

#### Inverse modelling and transport model performance

The performance of the inverse modelling approach depends critically on the quality of the atmospheric transport models that are used. Within the Bayesian framework, which is commonly used for GHG emission optimization, transport model uncertainties can formally be taken into account. Doing so reduces the weight of the measurements, which increases the uncertainty of the estimated (posterior) CO<sub>2</sub> fluxes. This means that transport model uncertainties reduce the precision of the estimated fluxes and thereby the performance of the inverse modelling approach.

The difficulty is that transport model errors are usually poorly quantified. Inaccuracies in the representation of transport model uncertainties in the inversion cause the estimated uncertainty to deviate from the true uncertainty. Furthermore, systematic errors and biases in transport models affect not only the precision but also the accuracy of the inversion-derived fluxes in a way that is difficult to quantify.

Due to the complexity of transport models it is difficult to assess uncertainties by formal error propagation methods. A common approach to assess transport model uncertainty is by comparing the results of different models using the same boundary conditions (i.e. initial concentrations and surface fluxes). A next step is to quantify the impact of transport model uncertainty on inverse modelling estimated fluxes. Such experiments have been carried out and studied in the past for global models in the framework of TRANSCOM (Gurney, Law et al. 2003; Baker, Law et al. 2006). The results indicate that the impact of transport model uncertainties on the estimated CO<sub>2</sub> fluxes is similar in size to the posterior uncertainty of the fluxes, as derived from a single inversion which ignores the impact of transport model uncertainty.

For regional-scale inversions the second experiment has not been carried out yet, but intercomparisons of forward models suggest that transport model uncertainties are more critical at the regional scale compared with the global scale (Geels, Gloor et al. 2007). This is explained by the fact that at the regional scale, for example in Europe, the landscape consists of a complex pattern of sources and sinks, which leads to a high variability in the CO<sub>2</sub> concentration measurements. To resolve that variability puts stringent requirements on the spatial and temporal resolution of meso-scale models. This is confirmed by (Gerbig, Lin et al. 2006) who demonstrate that continuous CO<sub>2</sub>

measurements at a tall tower can be reproduced by a mesoscale model provided that the resolution is high enough (a few km<sup>2</sup>).

As part of the current ME2 project we have worked on several aspects of transport modelling, which are described in the following subsections. On one hand we worked on improving fundamental transport parameterisations in atmospheric transport models. On the other hand, we carried out two model intercomparison experiments to further investigate transport uncertainties at the regional scale. The first experiment was performed at low resolution (~50x50km<sup>2</sup>) for the European domain to verify the results of (Geels, Gloor et al. 2007). A second experiment was initiated at high resolution (~5x5 km<sup>2</sup>) for the domain of The Netherlands using the measurements collected within ME2 to further investigate the approach of (Gerbig, Lin et al. 2006). However, analysis has not yet been concluded and results are not part of this report. Finally, we participated in a model intercomparison organized within CarboEuropeIP, specifically targeting CO<sub>2</sub> emissions of fossil fuel combustion.

#### Parameterisations of fundamental transport characteristics

In order to infer source strengths of Greenhouse gasses (GHG's) from observed concentrations, the quality of the prescribed atmospheric transport should be of high quality. This applies both to the horizontal transport (advection) and to the vertical transport (turbulent diffusion and exchange with the free troposphere).

Horizontal advection in the boundary-layer depends on the wind field, which in turn depends on the large-scale forcing and vertical turbulent momentum transport. In order to better understand and predict the wind field during day-time use has been made of a mixed-layer model. In particular, we have studied the oscillatory behaviour of the wind speed and direction. It turns out that the properties of the oscillator are strongly dependent on the way surface friction is parameterized (Schröter, Moene et al. 2011 (subm)). For a boundary layer without entrainment, the damping and period of the oscillation are significantly larger when a quadratic drag-law is used (which is consistent with the neutral limit of Monin-Obukhov similarity) as compared to a linear drag law. On average the damping and oscillation period of the linear model are 2/3 and 1/2 of that of the quadratic model. In the limit of small amplitudes this can also be shown analytically. The addition of entrainment changes in particular the equilibrium velocity components (Schröter, Moene et al. 2011 (subm)). Those are not only time-dependent because the boundary layer depth differs from time to time (static equilibrium) but also due to the growth itself (dynamic equilibrium). Finally, it can be shown that even in a well-mixed convective boundary layer a wind-maximum can occur in the lower part of the boundary layer, due to the inertial oscillation.

In transport models the vertical transport of scalars is parameterized by the boundary-layer scheme. Previous studies indicate that night time mixing is often overestimated, that the low-level jet is misrepresented, and that the nocturnal surface cooling needs improvement (Steenefeld, Mauritsen et al. 2008). On the other hand, the representation of daytime ABL entrainment could be improved as well. The PBL schemes implemented in the mesoscale model WRF are evaluated against a network of in situ observations in The Netherlands. Figure 3.1 shows one example of the skill of three state-of-the-art boundary layer schemes with respect to the boundary layer profiles of wind. Neither the wind speed nor the wind direction in the boundary-layer is represented correctly. Figure 3.2 shows a validation of the models using some in-situ observations from the scintillometer / ceilometer network. It is striking that the surface sensible heat flux as simulated by the model is more than a factor of two off as compared to the observations. The fast initial boundary layer growth is best captured by the models. For more details see (Steenefeld, Hartogensis et al. 2008; Steenefeld, Tolck et al. 2011).



To further improve the understanding and representation of boundary-layer transport during stable conditions we have participated in the 3<sup>rd</sup> model intercomparison study in the context of GABLES, both for single column models and LES models (Basu, Steeneveld et al. 2008; Bosveld, Bruijn et al. 2008; Moene, Baas et al. 2011).

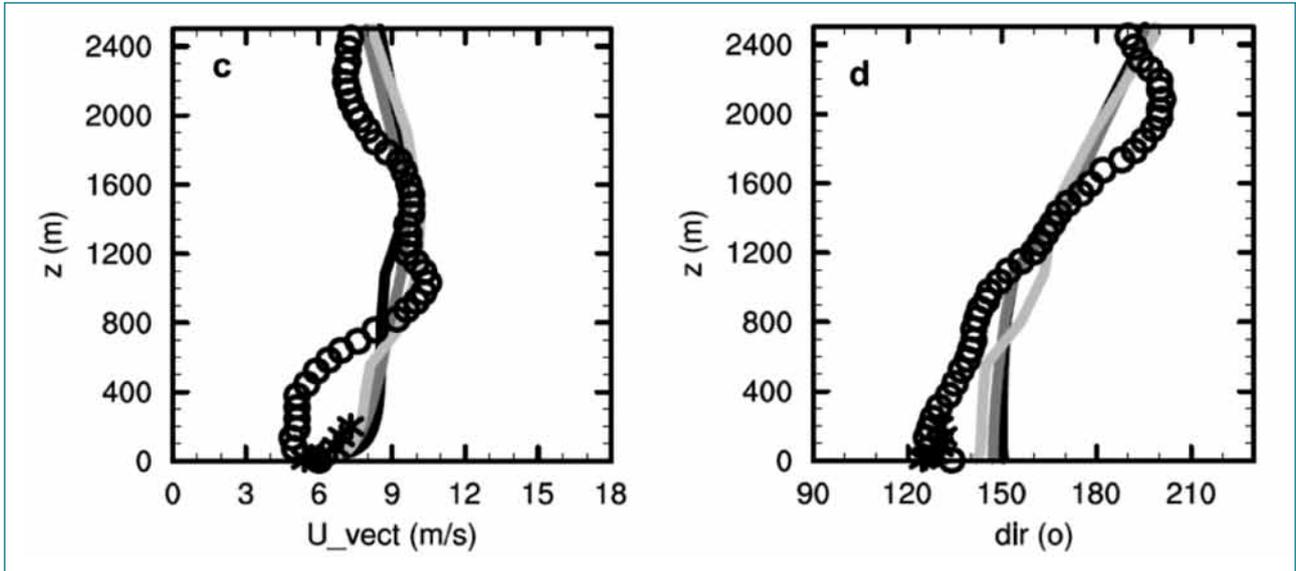


Figure 3.1. Modelled and wind speed (left) and direction (right) for 12 June 2006, 12 GMT. Asterisk: Cabauw observations, o= radio soundings. Boundary-layer parameterizations: YSU (black), MRF (medium grey), MYJ (light grey).

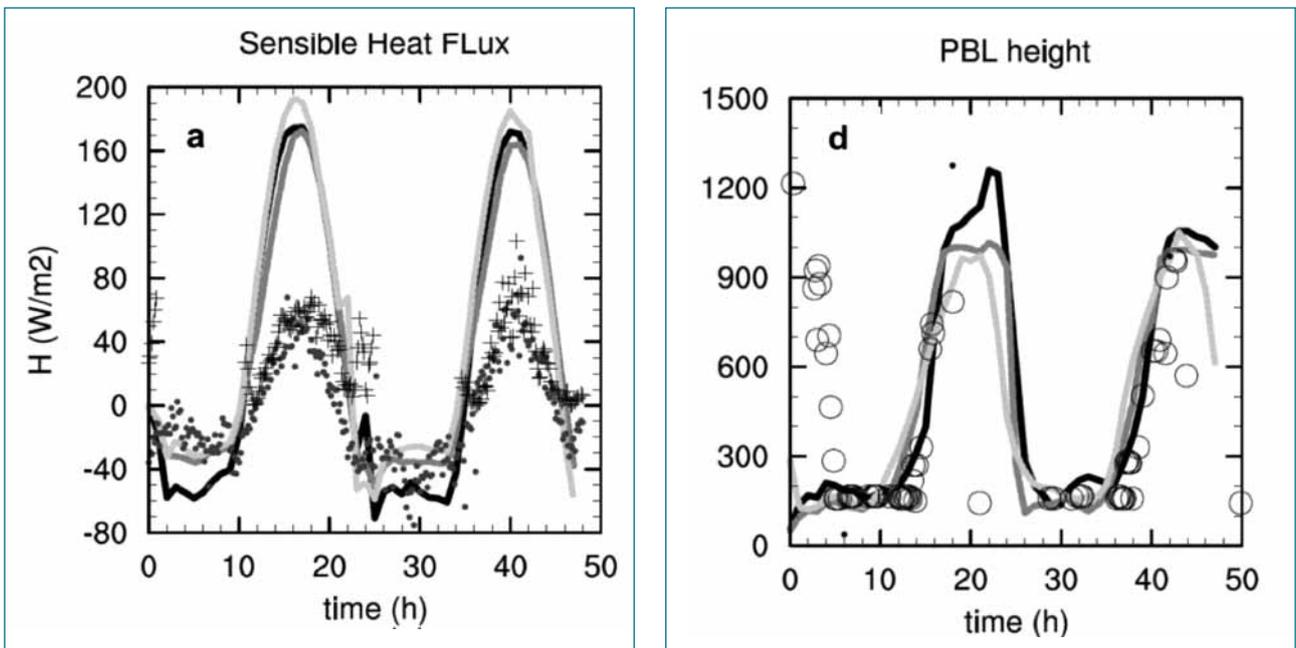


Figure 3.2. Modelled and observed sensible (left) and boundary-layer height (right). Dots: eddy-covariance, + scintillometer, o = ceilometer. Boundary-layer parameterizations: YSU (black), MRF (medium grey), MYJ (light grey). Time is hours after initialization (10 June 2006, 18 GMT)

Further analysis of this, and including also the RAMS model, learned that both WRF and RAMS overestimate friction velocity and near surface wind speed, even after adjustment of model parameters (Steenefeld, Tolk et al. 2011). An important inconsistency was found regarding the ABL daytime heat budget: Both model versions are only able to correctly forecast the ABL thermodynamic structure in case the modelled surface sensible heat flux is much larger than the observations indicate. Sensitivity studies and evaluation of radiative tendencies and entrainment reveal that possible errors in these variables cannot explain the apparent missing sensible heat flux within the current model infrastructure.

#### Assimilation of local observations

An accurate knowledge of the structure of the atmospheric boundary layer becomes important when trying to discern GHG sources and sinks from atmospheric concentration observation. This is especially the case when estimates are needed on increasing spatial resolutions. At high resolutions the transport from the sources/sinks to the receptor point cannot be approximated by a fully mixed atmospheric boundary layer flow. We developed a method to estimate the structure of the boundary layer given limited observations. A data-assimilation system has been set-up, which combines the observations with a state-of-the-art atmospheric model. At KNMI, a Regional Atmospheric Climate Model (RACMO) (Meijgaard, van Ulft et al. 2008) is run in forecast mode on a continuous basis. A Single Column Model (SCM) is directly derived from RACMO. From the 3D operational RACMO runs we derive dynamic forcings for the Cabauw column (Bosveld, Meijgaard et al. 2008). In search of an appropriate data-assimilation method first a variational technique was pursued on the relatively simple problem of the thermodynamics of the soil in Cabauw (Ronda and Bosveld 2009). Implementing this technique on the much more complex full atmosphere-soil model proved to be too complicated. Alternatively, the method of ensemble Kalman filter (enKF) was selected (Baas and Bosveld 2010).

Instead of using only one model realization, in an enKF system a collection of model realizations is used. Randomly disturbed initial conditions and large-scale forcings introduce small differences in the various model realizations. The spread among the ensemble members represents the uncertainty in the model forecast. By comparing this uncertainty with the estimated observation errors, an optimal estimate of the state of the atmosphere can be derived.

As an example, Figure 3.3 shows the impact of the data-assimilation system for one month of SCM forecasts. In this case, near-surface observations of temperature, specific humidity and both components of the wind vector were assimilated. Figure x compares model results for simulations with (enKF) and without (Empty) assimilation of observations with soundings from De Bilt at 12 UTC. It demonstrates that the assimilation of near-surface observations significantly reduces the root mean square error in a deep layer of 1000 to 1500 m above the surface. Combining model information with observations appears to provide a more accurate estimation of the state of the lower part of the atmosphere than can be obtained from both sources separately.

The enKF system has been implemented in the KNMI test bed. Each hour a new forecast is made for which the initial conditions are composed of the 1 h forecast of the previous run and observations valid at the same time.

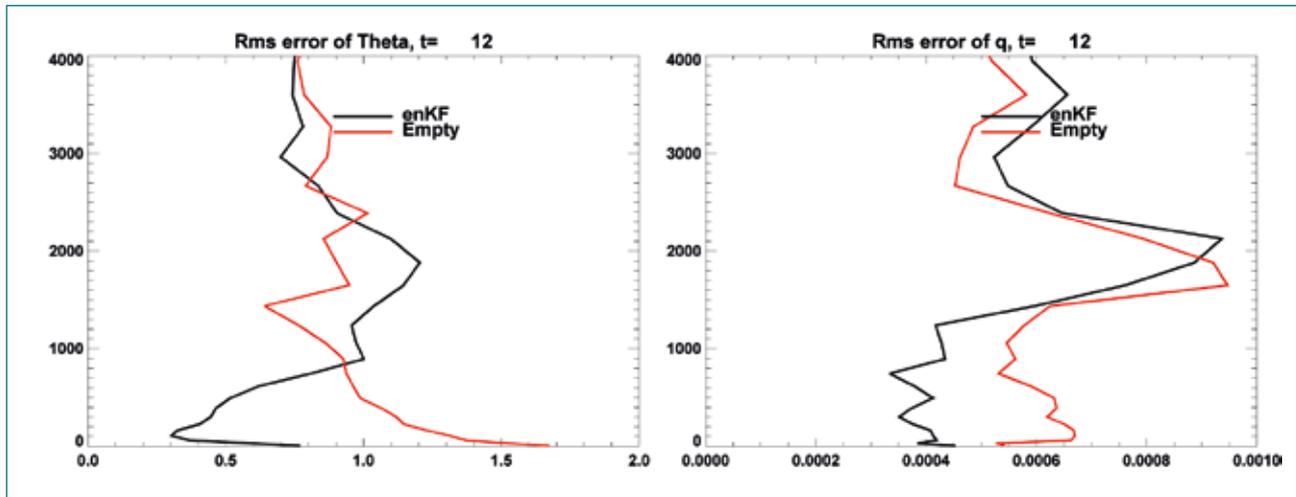


Figure 3.3.

RMS profiles of potential temperature (K) (left panel) and specific humidity (kg/kg) (right panel) at 12 UTC. Average values over 31 forecasts of the month of May 2008. The rms calculations are based on a comparison with soundings from De Bilt at 12 UTC. The black line presents the enKF results, the red line the Empty results.

#### Low-resolution CO<sub>2</sub> transport model intercomparison

This experiment is a modified version of the TRANSCOM continuous experiment (Law, Peters et al. 2008). It has been modified to allow contributions from regional scale models. For the specification of initial and lateral boundary conditions global concentration fields were made available derived from the TM5 model. Output was requested for 54 surface measurement sites, including a subset of 32 elevated sites for which vertical profiles were stored from the surface up to 500 hPa. The original station list of the TRANSCOM continuous experiment was extended with additional European measurement sites.

The analysis focused on the European domain for the period 2002-2003. Calculations were carried out for CO<sub>2</sub>, SF<sub>6</sub>, and <sup>222</sup>Rn. The CO<sub>2</sub> surface fluxes account for contributions from the ocean (Takahashi, Sutherland et al. 2002), fossil fuel use and cement production (Edgar 3.2 distribution scaled to CDIAC country data) and the terrestrial biosphere (CASA-GEFD2, (van der Werf, Randerson et al. 2006). CASA fluxes were extended with a simplified parameterization of the diurnal cycles of photosynthesis and respiration. These fluxes were made available at 3 hourly time resolution. SF<sub>6</sub> emissions are based on the EDGAR V3.2 inventory for 1995, scaled to match the global growth rate as observed by the NOAA-ESRL surface network. <sup>222</sup>Rn emissions were assumed to be constant using typical rates for land and ocean, including a highly simplified latitudinal dependence (see the experimental protocol for more detailed information).

Table 3.1 lists general characteristic of the 5 transport models which participated in the experiment. Most models are Eulerian regional domain models, except COMET (regional, lagrangian) and TM5 (global, eulerian).

Table 3.1.

Low resolution model intercomparison: Overview of participating models

Model	Domain	Hor. resolution	Vert. Resolution	Institute	Reference
TM5	Global	1°x1°	25 lev.	SRON/IMAU/ WU	Krol et al. (2005)
Comet	NH	0.5°x0.5°	30 lev.	ECN	(Vermeulen, Pieterse et al. 2006; Pieterse, Vermeulen et al. 2008)
RAMS-VU	Europe	80x80km <sup>2</sup>	39 lev.	VU-Falw	(Pielke, Cotton et al. 1992)
RAMS-Alt	Europe	48x48km <sup>2</sup>	34 lev.	Alterra	(Pielke, Cotton et al. 1992)
Lotos-Euros	Europe	0.5°x0.25°	4 lev. <3500m	TNO	(Schaap, Timmermans et al. 2008)

Figure 3.4 provides a few typical examples of results that are obtained from this experiment. Significant differences are found between the models, highlighting the common difficulty to realistically represent the night-time planetary boundary layer. Concentrating on daytime only, however, also significant differences remain (range = ; std = ; corr.coef= ). Generally, it is difficult to rate the relative performance of the models. Certain models score well at certain sites, but poorly at others. Even at a single site the performance varies from event to event. This is true also for the lower resolution global model, which doesn't show a systematically reduced performance in comparison with the regional models. It should be mentioned, however, that the underlying emission fields are available only at 1°x1° so that, in this experiment, regional models cannot take full advantage of increased resolution.

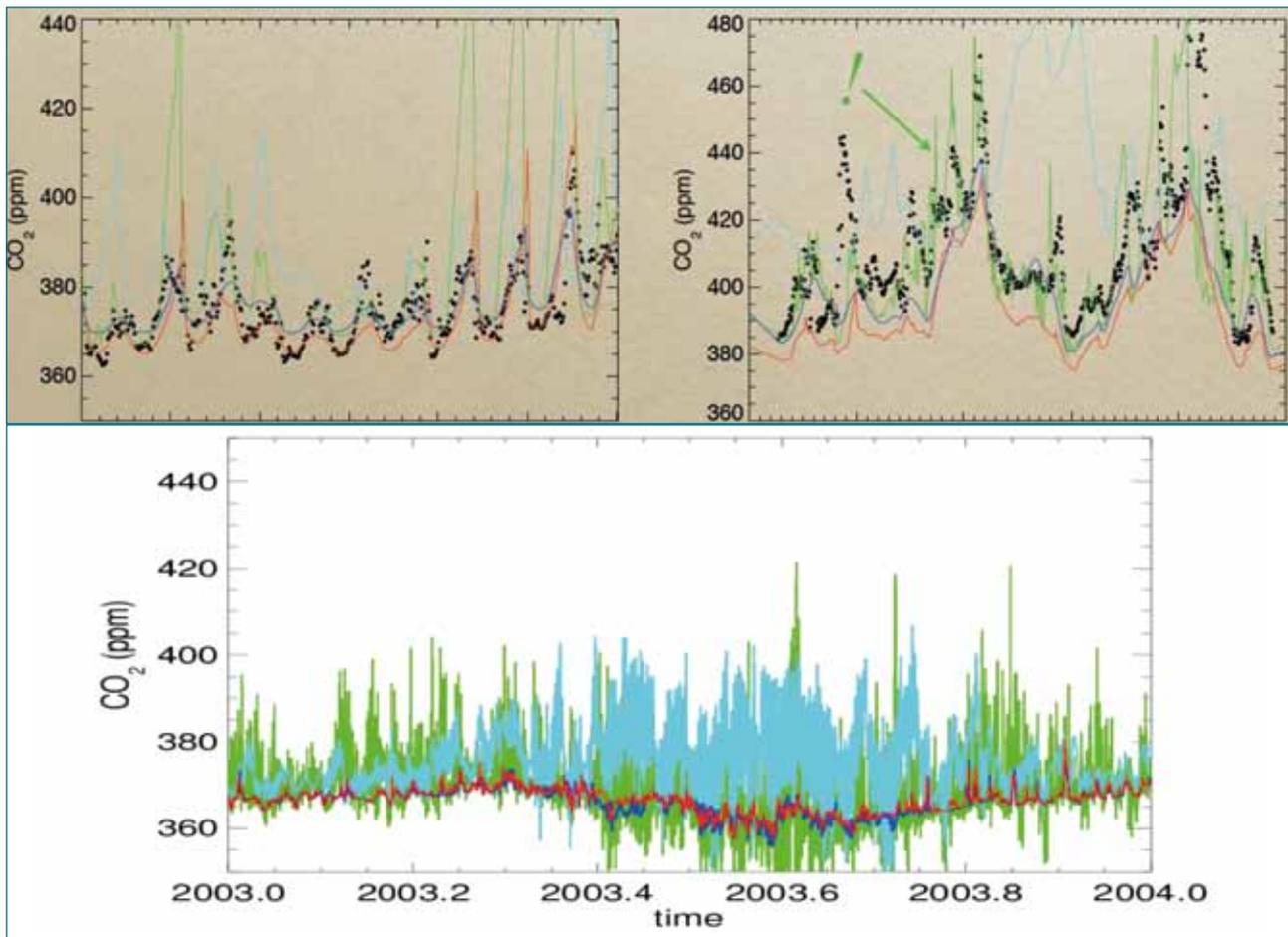


Figure 3.4.

Comparison of CO<sub>2</sub> concentrations measured in summer (Aug. 2003, left) and winter (Dec. 2003, right) at zoom at Cabauw (black dots) against those simulated by TM5 (red), COMET (green), RAMS-VU (dark blue), RAMS-Altterra (light blue) and LOTOS (magenta).

#### High-resolution model intercomparison

The high-resolution model intercomparison focuses on the domain of The Netherlands, for the period May-June 2008. The target period was limited to 2 months to facilitate model calculations at high resolution. The year 2008 was selected because it was designated ‘the golden year’ within the ME-2 project component, in which most of the measurement activities took place. The months May and June were selected for favourable measurement coverage and variable meteorological conditions.

Calculations were carried out for CO<sub>2</sub> and <sup>222</sup>Rn. In addition, planetary boundary layer heights were stored for comparison with available ceilometer measurements. The models were sampled at the coordinates of 22 flux and tall towers and 31 aircraft measurement transects including vertical profile measurements.

Surface flux fields were made available at a horizontal resolution of 0.10x0.10 and 3 hourly time resolution (biosphere only), using the FACEM biosphere model for biogenic CO<sub>2</sub> fluxes (Pieterse, Bleeker et al. 2007), EDGAR v4 fossil fuel fluxes including sector dependent diurnal, weekly and monthly variations (European Commission and (PBL) 2010), and Takahashi ocean fluxes (Takahashi, Sutherland et al. 2002). For an overview of the participating models see Table 3.2.

Table 3.2.

High resolution model intercomparison: Overview of participating models

Model	Domain	Hor. resolution	Vert. Resolution	Institute	Reference
TM5	Global			SRON/IMAU	Krol et al. (2005)
WRF	Europe			ECN	(Skamarock, Klemp et al. 2005)
VU(-RAMS)	Europe	5km	40 levels	VU	(Pielke, Cotton et al. 1992)
ALT(-RAMS)	Europe	2km	>50m 35 levels (12 in PBL)	Alterra	(Pielke, Cotton et al. 1992)

The analysis is still in progress. Tentative and yet incomplete results reveal a number of features.

Simulated CO<sub>2</sub> fluxes generally exhibit reduced diurnal ranges when compared to observed magnitudes. This applies strongest for Cabauw where simulated night-time fluxes are only half of the very high respiration fluxes (~10 umol.m<sup>-2</sup>.s<sup>-1</sup>, due to peat oxidation?) while daytime simulated uptake of the grassland is even less than half observed (-6 vs -17 umol.m<sup>-2</sup>.s<sup>-1</sup>). By comparison the night fluxes at Loobos pine forest are well simulated (~5 umol.m<sup>-2</sup>.s<sup>-1</sup>) but day-time uptake is only 60% of observed (-9 vs -15 umol.m<sup>-2</sup>.s<sup>-1</sup>). The Zeewolde simulated fluxes are simulated fairly constant over the two months between -5 in day and +1 umol.m<sup>-2</sup>.s<sup>-1</sup>, while in reality it is a bare field in the first month with small source of CO<sub>2</sub> (+2 umol.m<sup>-2</sup>.s<sup>-1</sup>) while after germination the maize on the site takes up over 30 umol.m<sup>-2</sup>.s<sup>-1</sup> towards the end of the second month.

Observed PBL heights vary over the country (see figure 3.5). Of the five sites monitored with the Vaisala ceilometer described in the previous chapter, *Deelen* on average exhibits the highest daytime maximum PBL at almost 1700m, probably due to its high sensible heat flux resulting from low albedo forests (high available energies) and low evaporation. Next *De Bilt* and surprisingly *Terschelling* on average reach about 1400m while *Beek* and *Eelde* reach only 1200m on average in these spring months. The models have difficulty reproducing these patterns. In daytime the coarsest resolution model (TM5) underestimates daytime PBL heights at all sites, while the highest resolution model (ALT) overestimates these. At night the VU model overestimates stable boundary layer height, TM5 and ALT underestimate it somewhat.

As a result, the models all have difficulty simulating the diurnal range in CO<sub>2</sub> concentrations at the elevated observation heights of 60m at Lutjewad and 200m of Cabauw. Overall the models reproduce better the 20m concentration dynamics (corr 0.45-0.70) than they do the 200m dynamics (corr 0.11- 0.44). See figure 3.6. The latter exhibits more short term variability at night and in the morning which seems to be related to the imprecise simulation of PBL heights close this observation height giving rise to fast fluctuations between low free troposphere concentrations and (very) high SBL concentrations. Especially the TM5 and VU models do not reproduce the high night-time concentrations that can be observed. While for VU this may be consistent with its overestimation of night-time PBL heights, TM5 has one of the shallowest stable boundary layers. Daytime observed minimum concentrations can in principle be reproduced by the models although the ALT has difficulty doing so, consistent with its generally too high CBLs. Night time peak concentrations generally come too early in the simulations by upto 2-3 hrs; daytime minima come too late by upto 3hrs. At the lowest levels of 20m at Cabauw all models do much better, both day and night.

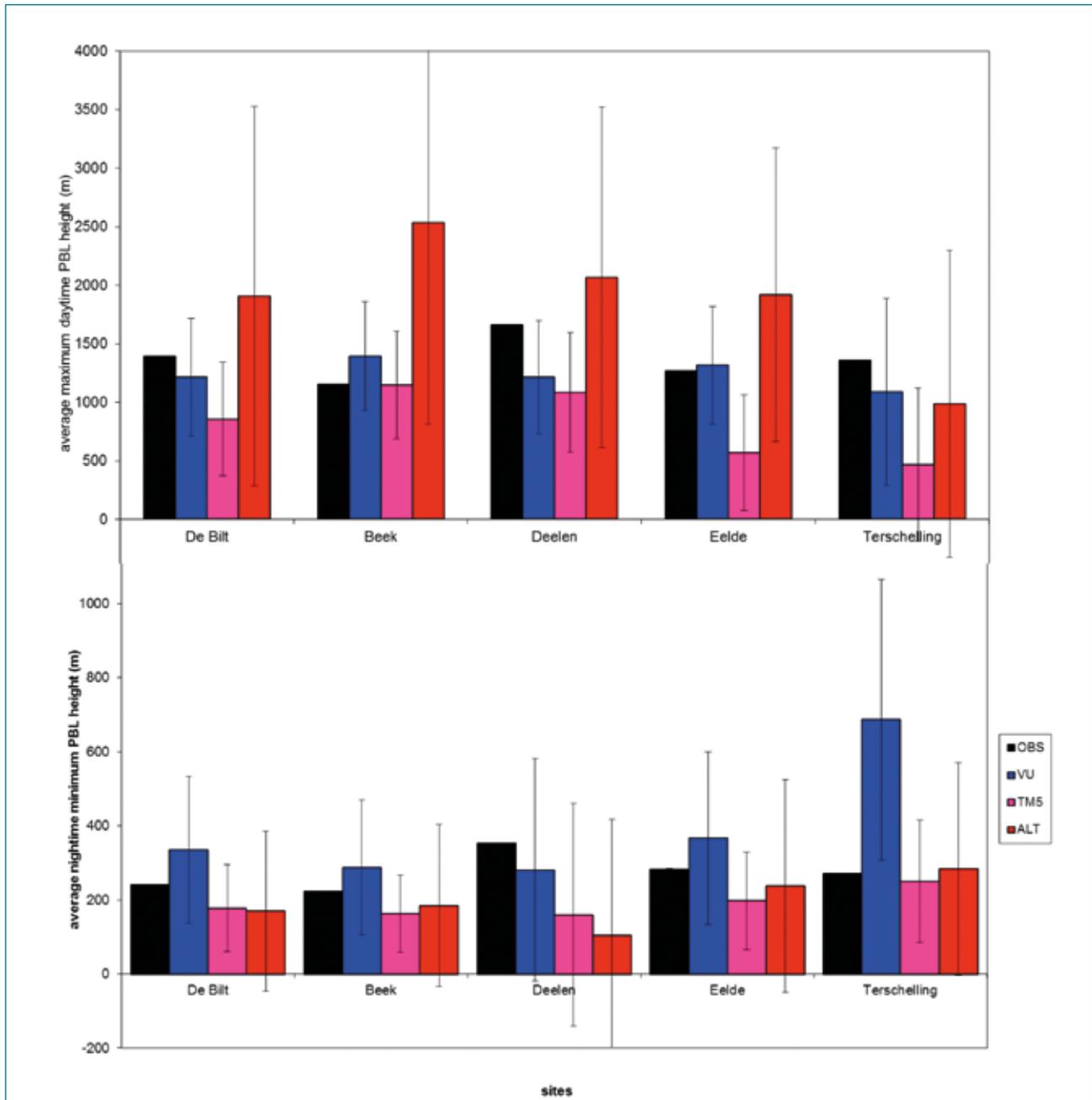


Figure 3.5. Observed and simulated PBL heights: daytime maximum (top) and night time minimum (bottom).

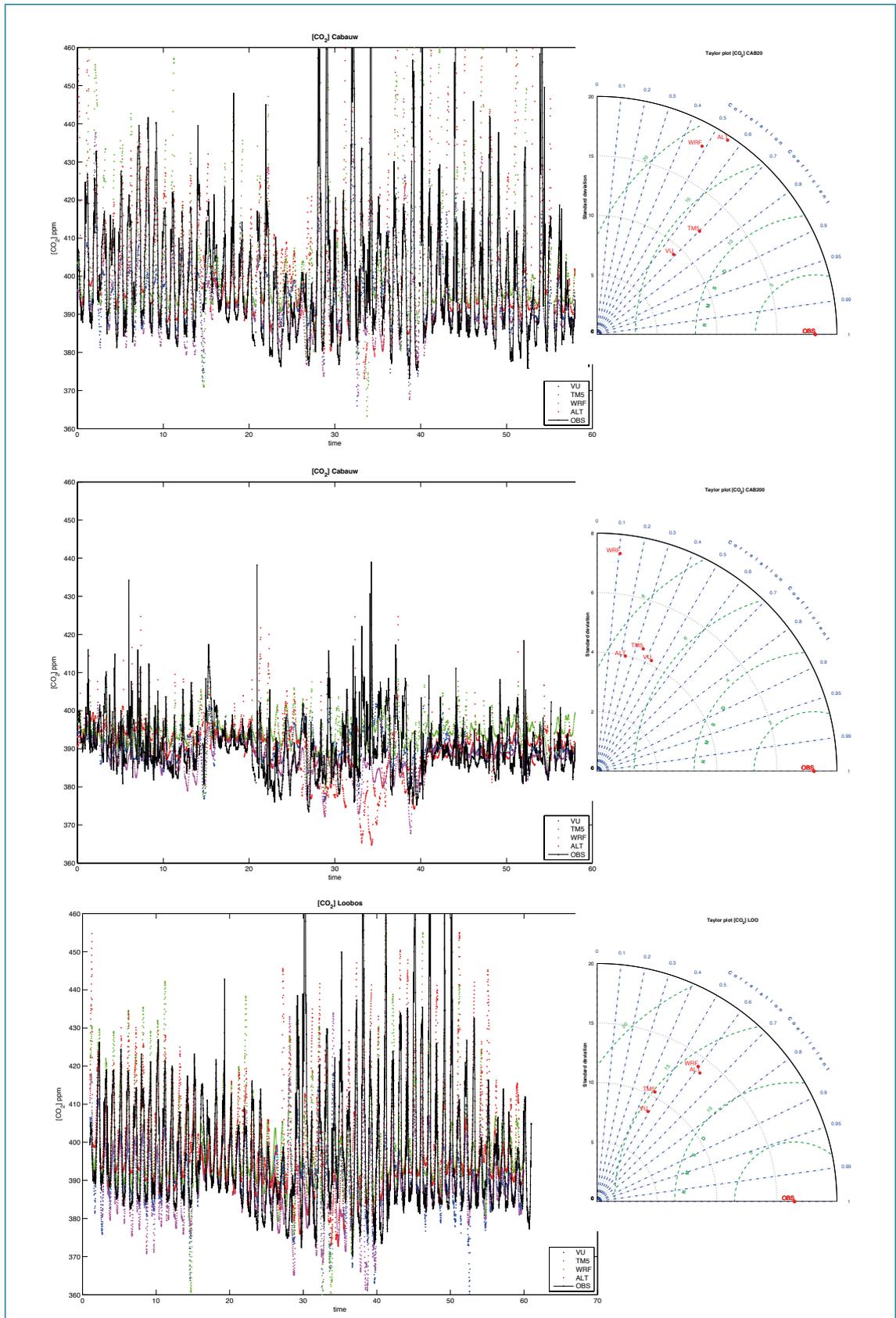


Figure 3.6. Simulated and observed CO<sub>2</sub> concentrations at the 20m level at Cabauw (top) at 200m at Cabauw (middle) and at 25m at the Loobos site. All time series plots have identical legends with VU – blue, TMS – pink, WRF – green, ALT – red and observations in black. Also the corresponding Taylor diagrams are shown.



### Fossil fuel model intercomparison

The aim of the fossil fuel model intercomparison is to assess the importance of the short-term variations of fossil CO<sub>2</sub> emissions. Examples are diurnal variations (rush hours), weekly variations (weekend versus working days) and seasonal variations (residential heating during winter). These variations are usually neglected in inverse modelling studies. The justification for this simplifying assumption is that the observed CO<sub>2</sub> variability over land is generally dominated by variations of the biosphere flux. The question, addressed in this experiment, is how important variations in fossil fuel emissions are in the case of Europe, where fossil fuel emissions are the dominant term in the annual carbon budget (Peylin, Houweling et al. 2009). Related is the question what could be the use of Europe wide <sup>14</sup>CO<sub>2</sub> measurements (or a combination of <sup>14</sup>CO<sub>2</sub> and CO measurements) as proposed by (Levin, Hammer et al. 2011). The primary goal is to detect trends in fossil fuel CO<sub>2</sub> emissions (the signal of radiocarbon free fossil fuel emissions in atmospheric <sup>14</sup>CO<sub>2</sub> is easily detected). On the other hand, fossil emissions are believed to be rather accurately known (<10% uncertainty). Then depending on the performance of atmospheric transport models <sup>14</sup>CO<sub>2</sub> could rather be a useful tracer for validating atmospheric transport models.

The model intercomparison was carried out using several global and regional transport models, including TM5. Alternative European fossil fuel emissions inventories were tested including EDGAR (European Commission and (PBL) 2010) and IER (<http://carboeurope.ier.uni-stuttgart.de>) with and without variability on the daily, weekly and seasonal time scale. The results show a sizeable impact of temporal variations in fossil fuel emissions on the simulated CO<sub>2</sub> concentration at moderately polluted sites. This translates into an important uncertainty because the available information is insufficient to accurately account for these temporal variations and how they vary regionally. The differences in annual emissions between IER and EDGAR were much larger than expected, exceeding 10% for several European countries.

The overall outcome of this study highlights the importance of uncertainties of fossil fuel emissions. <sup>14</sup>CO<sub>2</sub> measurements would provide useful information, although it would be difficult to derive accurate constraints on annual fossil fuel emissions given the importance of atmospheric transport model uncertainties and uncertainties in the intra-annual variation of fossil fuel emissions.

### Inverse modelling algorithms

In the context of the global and regional budgeting of greenhouse gases, inverse modelling refers to the estimation of surface fluxes of such gases using atmospheric transport models and in situ concentration measurements. Because of the large heterogeneity of the fluxes in space and time, which cannot be fully resolved by the measurements, a best guess is used as a starting point (referred to as a priori fluxes), which is improved by the inversion such that the agreement between model and the measurements is optimized. From the optimization statistics estimates can be obtained of the uncertainty of the inversion derived (a posteriori) fluxes. However, these uncertainties mostly reflect the internal consistency of the inversion set-up. Several sources of uncertainty, such as transport model uncertainty, are difficult to account for within the inversion and lead to underestimation of the real uncertainties. By using a variety of inversion set-ups and models, more realistic estimates of uncertainty can be obtained.

Several mathematical approaches exist for solving the surface flux optimization problem, which corresponds to minimizing a least squares cost function  $J$  of the form,

$$J(x) = (d - Hx)^T C_d^{-1} (d - Hx) + (x - x_{apr})^T C_x^{-1} (x - x_{apr}),$$

where  $d$  and  $x$  are vectors of, respectively, the atmospheric measurements and the unknown surface flux parameters, which are related through the model operator  $H$ . The subscript 'apr' denotes a prior information. The deviations from the measurements and the a prior flux parameters are weighted by the covariance matrices  $C_d$  and  $C_x$ . Several alternative formulations of the inversion problem are possible depending on the exact definition of the various terms in the cost function. For example,  $x$  can be some spatio-temporal discretization of the surface flux, either at the grid resolution of the model or aggregated into a set of ecoregions. Alternatively, in the case of a so-called carbon cycle data assimilation system (CCDAS),  $x$  represents uncertain parameters of an underlying process model incorporated in  $H$ . Vector  $d$  may contain various types of measurements from, for example, background flask sampling sites, tall towers, aircrafts, and satellites. A subset of those measurements, for example from measurement campaigns, is often left out of the inversion for validation of the inversion-optimized model.

In recent years the performance of inversions is improved by increasing the model resolution and by the increased number of available measurements (satellites!). This led to the development of highly efficient inversion algorithms, such as the variational approach (4D-VAR) and the Ensemble Kalman filter (Houtekamer and Mitchell 1998; Whitaker and Hamill 2002; Lorenc 2003), for dealing with large optimization problems. Alternatively the size of the domain is limited to (sub)continents or even individual countries (Rivier, Peylin et al. 2010; Schuh, Denning et al. 2010). Since the concentration variations inside such regional domains may be significantly influenced by long-range transport, regional scale inverse modelling requires boundary conditions, which are usually derived from global models. Alternatively, a global model can be used with regionally varying resolution (so-called zoom models).

In the case of carbon dioxide much of the observed signal, i.e. concentration variations in time and space, is caused by atmospheric transport, which puts stringent requirements on the accuracy of such models. The remaining variability can be attributed to sources and sinks of  $\text{CO}_2$ , which – at distance of centres of urban activity – is dominated by biospheric photosynthesis and respiration processes.  $\text{CO}_2$  measurements themselves do not allow a distinction between fluxes of anthropogenic or biospheric origin. However, since the a priori anthropogenic fluxes, as derived from statistical emission inventories, have much lower uncertainties, the inverse modelling-derived flux constraints mostly address the biosphere. This is different, however, for highly populated countries, such as the Netherlands where most measurement sites are significantly influenced by emissions from fossil fuel use.

The measurements from Cabauw and Lutfjewad show a large variability of up to ~100 ppm, much of which is explained by the diurnal dynamics of the planetary boundary layer. Therefore, what is needed from an inverse modelling perspective is a high horizontal resolution in combination with a realistic representation of boundary layer dynamics. The model inter-comparison experiments (see previous section), conducted in the framework of this project, highlight the need for an improved model representation of the planetary boundary layer.



## Continental inversions

### Estimation of European CO<sub>2</sub> emissions

European sources and sinks of CO<sub>2</sub> have been estimated for the period 2001-2007 using atmospheric inverse modelling on the basis of background measurements in combination with continuous CO<sub>2</sub> measurements from the European tall tower network (Peters, Jacobson et al. 2007; Peters, Krol et al. 2010). The inverse modelling system makes use of the Ensemble Kalman filtering technique applied to the TM5 model, which, for this application, zoomed in on Europe. Weekly fluxes have been estimated for 18 ecosystems spread across Europe. It is found that the average net ecosystem exchange of Europe for the period 2001-2007 amounts to 165 TgC.yr<sup>-1</sup>. A predominant contribution to this uptake is attributed to forests of Eastern Europe. European croplands appear to be a modest source of Carbon, although its contribution is difficult to separate from fossil fuel emissions and is therefore less robust.

Inversion-derived flux variations over the 8 year period show an increased CO<sub>2</sub> outgassing of 0.15PgC/yr for 2003, associated with the extreme summer drought which struck several European countries during that year. Besides 2003, a large negative NEE anomaly (from the perspective of the atmosphere) is found for 2005, which is attributed to favourable summer growth conditions during a strong negative phase of the North Atlantic Oscillation.

### Estimation of European CH<sub>4</sub> and N<sub>2</sub>O emissions

The results of (spatial) high resolution inversions of the average emission fluxes for methane and nitrous oxide using the COMET inverse model have been obtained for the years 2006 and 2007, as these are the first two years for which this first and unique dataset of well-connected continuous observations of concentrations is available. The observations for the years 2006+2007 consist of observations from the CHIOTTO/CE-IP network of tall towers, the RAMCES network (F), the UBA network (D), and the NOAA CMDL cooperative flask network. Where available the continuous data were used in hourly resolution.

<sup>222</sup>Rn observations can be used to calibrate and test the (vertical) transport schemes of the models but also to infer model-free emission estimates that can be confronted with the model based inversion results. Also for <sup>222</sup>Rn more observation stations have recently become available. The latest results will be used to infer the prospects for future work using even more dense networks.

By correlating events with high correlation of simultaneous mixing ratio increases of Rn and a tracer observed at CBW station and assuming a known constant Rn emission 'model-free' emission estimates can be performed. We plan to use the COMET footprint model here to better estimate the local Rn emission to correct the direct flux method. (See figure 3.7).

COMET is a relatively simple two layer Lagrangian trajectory model (Vermeulen et al, 2006) describing concentrations in mixed layer and residual layer along trajectory path. COMET is driven trajectory data calculated by the FLEXTRA model (Stohl et al, 2004) using ECMWF analysed wind fields.

The Source Receptor Matrices derived with COMET are used in a direct SVD inversion using an Iterative Source Aggregation Scheme (ISAS). In ISAS the SRM matrix is accumulated from the SRM's for multiple receptors, regularized by joining adjacent cells in blocks of 2x2, 4x4 etc, until all joined blocks have contributions of at least a given value relative to the maximum value in the initial SRM. In further steps blocks with uncertain derived emissions are removed, blocks that form dipoles are joined and blocks with associated small eigenvalues are removed from the equation. Removed

blocks are assigned the prior emission. This way only emissions of areas for which the model can determine emissions with a prescribed uncertainty (here 30%) are calculated in the method, the remaining areas are treated as if they have the prior estimate of their emission.

Methane and nitrous oxide global background data have been calculated using the TM5 model by JRC in 2x3 degree resolution. Prior emissions are used according to the NEU 6.2 protocol with a resolution of 1x1 degree. The resulting inverse estimated emissions compared to the prior emissions are shown in figure 3.8. Table 3.3 shows the resulting emission estimates per country compared to the prior (JRC estimate) of emission and the value derived by the Rn tracer method. It shows good agreements of N<sub>2</sub>O emissions for some countries (NL, B, L) but much higher (50-170%) inverse estimates for others (D, F, and UK).

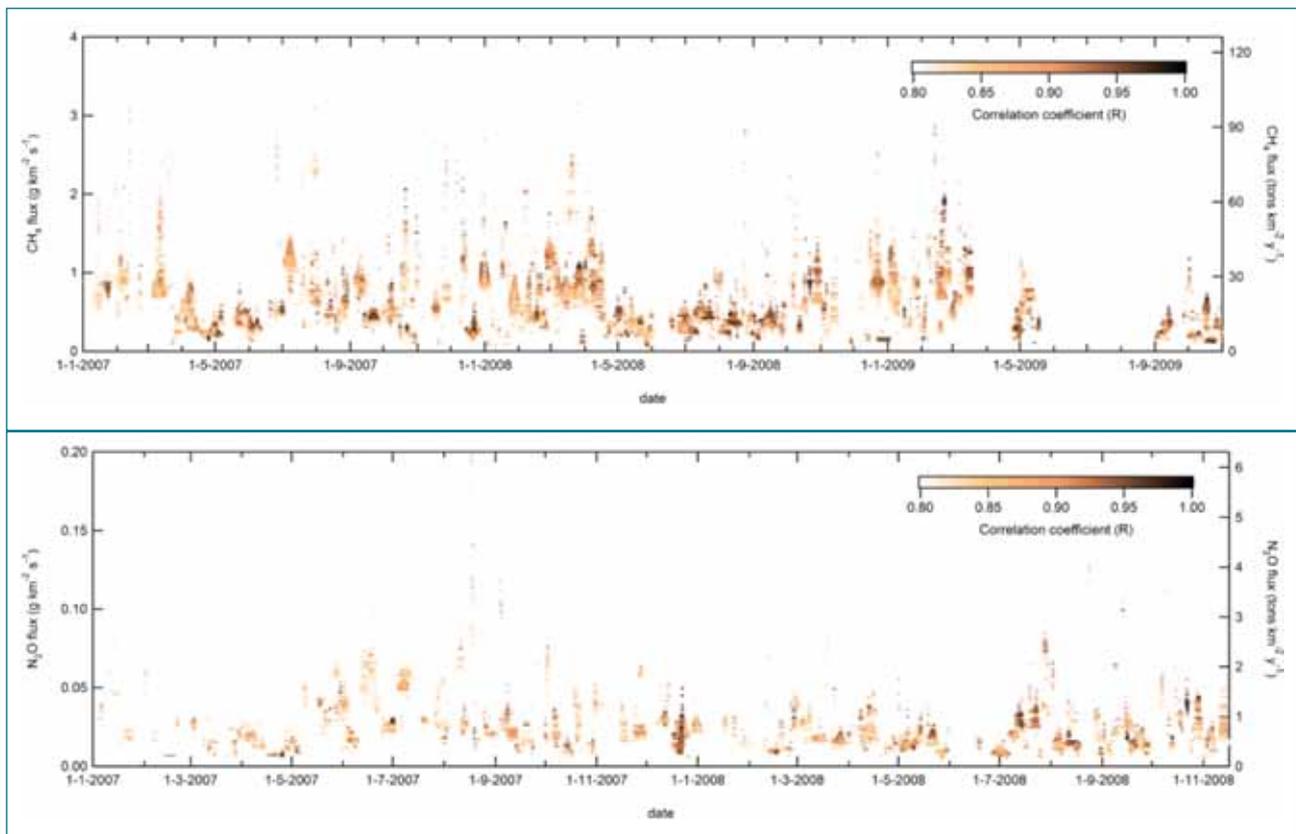


Figure 3.7.

CH<sub>4</sub> en N<sub>2</sub>O fluxes estimated with the direct <sup>222</sup>Rn flux method per pollution correlation event at Cabauw tower.

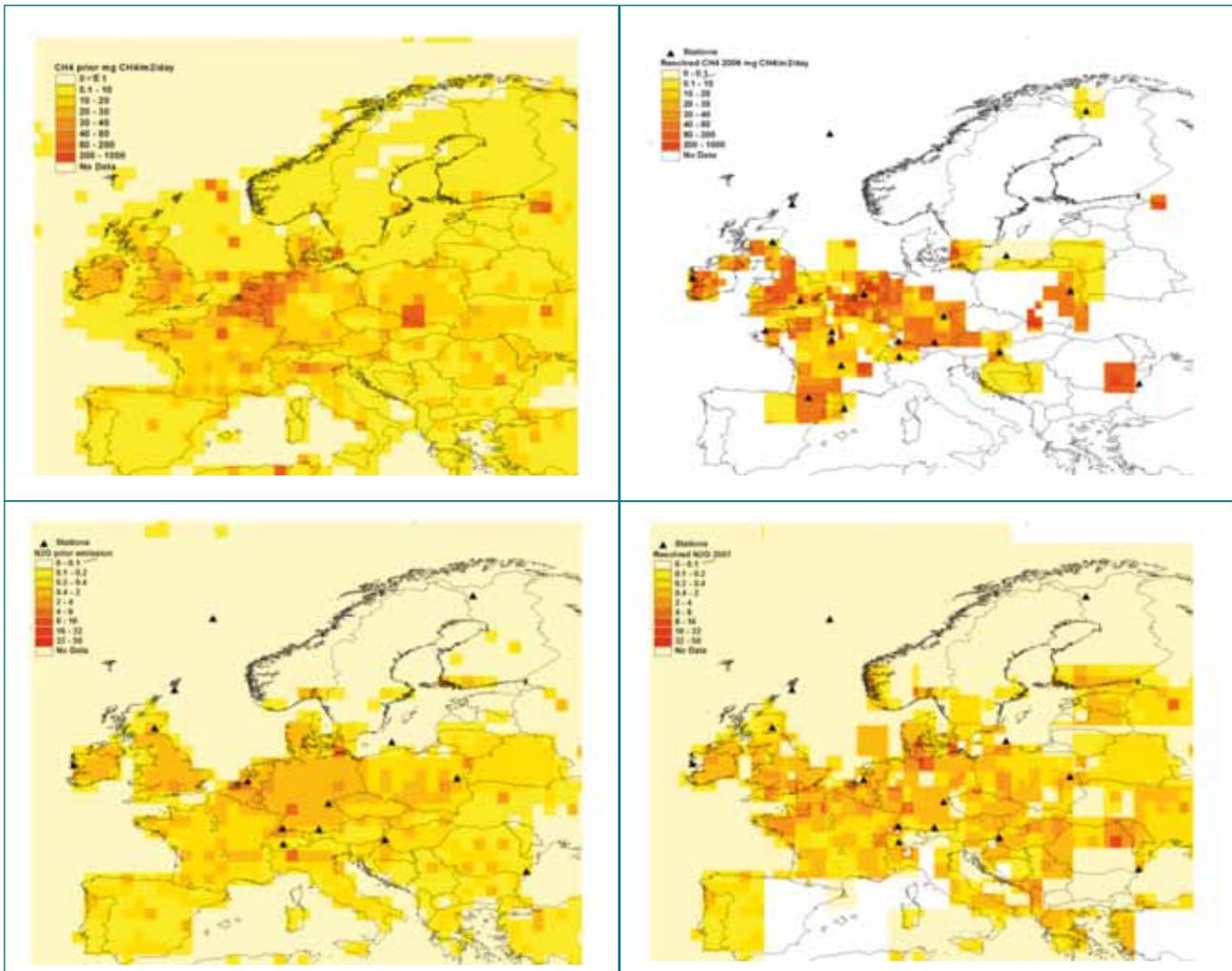


Figure 3.8.

Prior (left) and inverse resolved (right) emissions of CH<sub>4</sub> (top) and N<sub>2</sub>O (bottom), using the COMET inverse model and continuous observations of concentrations of CH<sub>4</sub> and N<sub>2</sub>O at the stations indicated with the triangles.

Table 3.3.

TM5 Emission estimates per country compared to the prior (JRC estimate) of emission and the value derived by the Rn tracer method.

2007	N2O			CH4		
	prior	resolved	Rn-method	prior	resolved	Rn-method
NL	1.24	1.226	1.84	58.2	64.8	46.2
B	1.52	1.53		58.2	67.4	
F	0.45	0.9		19.6	32.5	
D	0.84	1.3		22.1	52	
UK	0.54	1.46		18.9	38.6	
L	0.96	0.97		36.7	35.2	

mg/m<sup>2</sup>/day

## Regional scale inversions: towards estimation of Dutch GHG emissions

### Developing the forward model framework

Estimation of CO<sub>2</sub> fluxes for the Netherlands requires a higher spatial resolution than that applied to Europe in the previous section. Thereto, the Regional Atmospheric Mesoscale Modelling System (RAMS) has been equipped with surface exchange, and subsequent atmospheric transport of CO<sub>2</sub>. The RAMS model was used to simulate the domain of Western Europe, with a focus on the Netherlands where the model resolution was as high as 8km. Boundary conditions for this nested model simulation were taken from optimized CarbonTracker model results.

The setup of the RAMS model followed that described in the regional model intercomparison activity (Section 3.2), with the exception that (a) fossil fuels were prescribed at 8x8km resolution using the IER Stuttgart emission inventory (<http://carboeurope.ier.uni-stuttgart.de>), and (b) biospheric exchange was calculated in the model itself based on the LEAF land-surface model (Walko, Band et al. 2000) augmented with carbon dioxide exchange. The latter makes the calculated carbon balance consistent with the RAMS modelled surface energy and moisture balance.

Considerable effort was put into assessing this model against available BSIK observations from all over the Netherlands in the period May/June 2008. This assessment included 2m temperature, moisture, and wind speed/direction, the surface energy balance over forests, grass, and crops, the planetary boundary layer height, vertical profiles of temperature and moisture, net and gross carbon fluxes, and atmospheric CO<sub>2</sub> mixing ratios. The results were published in (Tolk, Peters et al. 2009) and is one of the most comprehensive studies of the exchange of heat+moisture+CO<sub>2</sub> over the country. An interesting finding from this work is that it was impossible to fit the observed vertical structure of the atmosphere while at the same time matching the observed surface energy balance (latent and sensible heat). Also, the stable boundary layer at night was insufficient well simulated to allow interpretation of night-time CO<sub>2</sub> mixing ratio values, as was found in other studies previously. Finally, this study presented an innovative analysis of carbon source signals in the measured CO<sub>2</sub> mixing ratios at Cabauw, illustrating the close ties between the observational and numerical groups in the BSIK consortium.

### Developing the inverse framework

Results from the (Tolk, Peters et al. 2009) study were promising enough to continue developing an inverse modelling framework. We chose to use the same method as in the CarbonTracker Europe parent system: ensemble Kalman filtering. However, the division of the domain into eco-regions, and subsequent scaling of NEE did not seem a logical choice for this much smaller domain. Therefore, in (Tolk, Dolman et al. 2011) we investigated 5 alternative approaches to optimizing the biospheric surface fluxes, each loosely based on existing frameworks in the field. The aim in this study was to correctly retrieve the FACEM fluxes used in Section 3.3, but now starting from a different first-guess biosphere model, and using a limited (but large) set of observations from the existing platforms in the Netherlands.

Our results showed that indeed, the scaling of NEE is a poor method for regional fluxes, while methods that optimize fluxes on a pixel-by-pixel basis or by estimating parameters of an underlying biosphere model performed better. Furthermore, this study brought to light a number of new issues specific to the regional inversions on higher resolution: (i) the importance of the underlying model structure in determining the outcome, (ii) the challenge of dealing with non-linear model formulations that appear unavoidable on these scales, and (iii) the need to anchor the regional dense network observations with 'integrating' measurement sites further downwind of the direct



sources and sinks. These considerations were taken into account for the 'true' regional flux estimates that are described next.

#### Estimating CO<sub>2</sub> exchange for the Netherlands

The inversion methods are being applied to obtain flux estimates for the Netherlands. Two of the methods compared in (Tolk, Dolman et al. 2011) are used: one in which the unknown biotic parameters are constant within an ecoregion, and one in which they vary from pixel to pixel. The first method assumes an unwarranted uniformity over the ecoregion, whereas the second method suffers from the high number of unknowns.

The method is applied for Spring, Summer, Autumn and Winter separately. However, winter fluxes are hard to monitor, because of their small magnitude and of the prevailing lack of convection which makes the transport hard to model. Only concentration measurements from 10 to 16 UT are used, since the transport model is less reliable for nocturnal hours. Data from Cabauw, Loobos, Lutjewad and Hengelman are used.

It is found that with prior values of the unknown parameters, the concentration can be already roughly reproduced most of the time. Since the differences are close to the uncertainty ( $\geq 5$  ppm for hourly values, with transport errors etc. included), it would seem difficult to obtain significant improvement of the parameters by inversions (Tolk, Peters et al. 2009; Tolk, Dolman et al. 2011). Indeed, the spatiotemporal patterns of the posterior fluxes depend rather strongly on the chosen inversion method. However, when aggregated over larger regions (say, province size), the prior-to-posterior change is often comparable at least for well-observed regions. Moreover, a comparison to fluxes observed by aircraft shows that posterior fluxes are (for most cases) much better than prior fluxes, once they are averaged over the flight trajectory. This holds for both inversion methods. For Winter, the inversion yields very little improvement, as was to be expected.

An investigation was done concerning the quantification of representation errors (difference between concentration at a point, and the smoothed average that models have to use because of their limited horizontal resolution). This was done for SW France, where the information from the CERES campaign could be used, but the results have a more universal applicability. The results were published in (Tolk, Meesters et al. 2008). It appears that at daytime, significant representation errors are caused by convective structures, mesoscale circulations (e.g. sea-breeze), as well as variability of the surface CO<sub>2</sub> flux. The meteorology appears the most important driver. At night, unresolved orographic heterogeneity is an important source of representation errors. Typical errors of this kind are 0.5 to 1.5 ppm for 20 to 100 km resolution. Larger errors should be expected however in the presence of fronts.

In (Meesters, Tolk et al. 2012 (subm)) inverse model results are compared to direct CO<sub>2</sub> flux measurements by aircraft, for 6 flight tracks over the Netherlands that were all flown multiple times in each season. After averaging over each trajectory for each season, posterior fluxes for summer and autumn are much closer to the observations than the priors, with a comparable performance for both inversion methods (pixel based and eco-region based resp.). The inversions, validated with the aircraft data, showed that the CO<sub>2</sub> fluxes in the Dutch region are more negative than suggested by the priors. This shows that the Netherlands is in 2008 a stronger sink than previously obtained from the biosphere model optimized with Fluxnet data.

### Data inversions for CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O over the Netherlands

(van der Laan, Neubert et al. 2009) present net emission estimates of CH<sub>4</sub> and N<sub>2</sub>O of The Netherlands based on measurements conducted during the period of May 2006 to April 2009 at station Lutjewad, The Netherlands (6021'E, 53024'N, 1m a.s.l.). 222Radon mixing ratios were applied as an indicator for vertical mixing and long-range air mass transport and used to calculate the net surface fluxes from atmospheric mixing ratios of CH<sub>4</sub> and N<sub>2</sub>O. Our study shows that our measurement site Lutjewad is well-suited to measure emissions from The Netherlands and validation of the national inventories using the 222Radon flux method. Since this study is purely observation-based it is independent from inventories or atmospheric models. Our results are compared to the national inventories as reported to the UNFCCC. They initially found (van der Laan, Neubert et al. 2009) net emissions of:  $(15.2 \pm 5.3)$  t km<sup>-2</sup> yr<sup>-1</sup> for CH<sub>4</sub> and  $(0.9 \pm 0.3)$  t km<sup>-2</sup> yr<sup>-1</sup> for N<sub>2</sub>O, later (van de Laan, 2010) with a modified procedure corrected to  $22.3 \pm 5.6$  t km<sup>-2</sup> yr<sup>-1</sup> for CH<sub>4</sub> and  $(1.5 \pm 0.4)$  t km<sup>-2</sup> yr<sup>-1</sup> for N<sub>2</sub>O. These latter values are similar to the inventory-based emissions (2006-2008 averages) of  $(18.3 \pm 3.3)$  t km<sup>-2</sup> yr<sup>-1</sup> for CH<sub>4</sub> and  $(1.3 \pm 0.6)$  t km<sup>-2</sup> yr<sup>-1</sup> for N<sub>2</sub>O, but the differences are insignificant.

Surface emissions of CO<sub>2</sub> from fossil fuel combustion (CO<sub>2</sub>FF) are estimated in (Van der Laan, Karstens et al. 2010) for the Netherlands for the period of May 2006–June 2009 using ambient atmospheric observations taken at station Lutjewad in the Netherlands. Measurements of Δ<sup>14</sup>C on 2-weekly integrations of CO<sub>2</sub> and CO mixing ratios are combined to construct a quasi-continuous proxy record from which surface fluxes are determined using the <sup>222</sup>Rn flux method. The trajectories of the air masses are analysed to determine emissions, which are representative for the Netherlands. We compared our observationally based estimates to the national inventories and we evaluated our methodology using the regional atmospheric transport model REMO. Based on 3 yrs of observations we find annual mean CO<sub>2</sub>FF emissions of  $(4.7 \pm 1.6)$  kt km<sup>-2</sup> yr<sup>-1</sup> which is in very good agreement with the Dutch inventories of  $(4.5 \pm 0.2)$  kt km<sup>-2</sup> yr<sup>-1</sup> (average of 2006–2008).

## 4. Can we verify reported emissions ?

### Conclusions present study

Over the past couple of years much progress has been made in terms of monitoring and modelling of GHG emissions in the context of verification. From the present project and its results as presented in this report and underlying publications a number of conclusions and recommendations can be made.

On the monitoring side the following conclusions and recommendations can be made:

- For the main GHGs continuous monitoring has become increasingly important as opposed to the dominance of flask sampling until not so long ago; especially for (sub)continental inversions.
- While until very recently measuring systems were relatively complex and to some extent custom build, and therefore required relatively large calibration and maintenance efforts, it has been proven that continuous datasets covering many years at high temporal resolution and high precision can be produced.



- Very recent instrument developments, however, resulted in much more robust high precision observations at much less maintenance costs. Especially cavity ring down techniques, presently commercially available for CO<sub>2</sub>, CH<sub>4</sub>, CO and some isotopes ( $\Delta^{13}\text{C}$ ) through Picarro, have become de-facto standards for future monitoring efforts.
- Despite the current practice to use only a fraction of these continuous data streams, filtering out periods where transport models have still major difficulties (e.g. night time), or periods in which the signal is dominated by unwanted areas (near/far field) depending on the objective of the study, we strongly recommend to continue such continuous observations. The filtering needs will very likely change in the (near) future and having in place a long term data set will then be invaluable
- Flask sampling remains important for difficult to analyse gases and especially isotopes like <sup>14</sup>C important to discriminate between fossil and present sources of CO<sub>2</sub> and other (clumped) isotopes like <sup>17/18</sup>O.
- The tall tower concept and use, in the EU led by Dutch initiatives, has now been well established and a network of such towers is operational and being extended. Their height places them well into the mixed layer giving them a large footprint and a signal little or occasionally influenced by very local sources and sinks.
- Some tall towers, like in Cabauw, sample the GHG profile at multiple levels. Though also here at present only the top level data are most used in inversions studies, data from lower levels will become more important as the ability of our atmospheric models to represent these increases. Again, at that time having available a long term data set will be invaluable.
- National inversion estimates will be better constrained when the present network will be extended with a number of towers around the borders of the country. Unpublished synthetic studies (Vermeulen, 2008, Tolk 2008 pers. comm.) have demonstrated for both CO<sub>2</sub> and CH<sub>4</sub> the value of stations along the south and eastern border (better constraining inflow from these directions) as well at sea (back ground signal).
- For the interpretation of GHG concentrations measured inside the PBL an estimates of its height is invaluable. We have developed and established a first ever network of PBL monitoring stations using ceilometers. Their use is now well recognised; and the deployment of ceilometers is now part of standard ICOS Level 1 Atmosphere stations.
- Estimates of regional fluxes can be made directly using aircraft observations of fluxes. The technique and processing efficiency have been progressed in the present project to levels allowing for operational measurements, covering large areas and full seasonal cycles.
- Despite progress gained on the retrieval of CO<sub>2</sub> from SCIAMACHY over the years, it has not reached the accuracy level needed for global source and sink estimation, mostly due to difficulties to account for cirrus and aerosol scattering. For CH<sub>4</sub> the situation is much more favourable due to the so-called proxy approach which cancels out errors due to aerosol and cirrus scattering cancel out to a large extend.

Similarly a number of conclusions and recommendations can be made with respect to transport modelling and inversion algorithms

- Significant differences are found between 3D transport models, highlighting the common difficulty to realistically represent the night-time planetary boundary layer. Also significant differences remain in daytime. Generally, it is difficult the rate the relative performance of the models, as model performance varies across sites and between events.
- Thus, there is still considerable room and need for improvement of the modelling of PBL dynamics in mesoscale models. No single scheme performs consistently better than alternatives in all situations. Vertical mixing (especially in stable conditions) but also wind speed and direction need better representation.

- Part of the misrepresentation of PBL dynamics is reflected in the fact that the simulated surface energy balance does not match with atmospheric observations when applying well established modelling techniques. A paradox that needs to be resolved.
- Until such issues have been resolved it may be advisable to use an ensemble of transport models.
- Data inversions of all three major GHG has been proven feasible, using  $^{222}\text{Rn}$  as a transport tracer. Rather directly this resulted in good estimates for  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , probably due to their relative homogeneous and diffuse source distribution in the footprint of Lutjewad tower.
- Data inversions for fossil fuel derived  $\text{CO}_2$  are feasible when discriminating it from biogenic  $\text{CO}_2$  using continuous  $\text{CO}$  and event sampling of  $^{14}\text{C}$ . Yet more transport information was needed than for  $\text{CH}_4/\text{N}_2\text{O}$  in order to distinguish between near field low emissions and far field high emissions.
- The signal in  $\text{CO}_2$  variations at Cabauw is dominated by the fluxes in its footprint, not by transport errors, a pre requisite for successful model based  $\text{CO}_2$  inversions. But also the signal variability in total  $\text{CO}_2$  at Cabauw seems dominated by biospheric fluxes despite the proximity of important urban centres.
- Considerable progress has been made in pixel based model inversions, across a wide range of scales, using 4DVAR and recursive source area aggregation SVD inversion techniques. Pixel based parameter optimisations ranged from  $1^\circ$  resolution at the Global/European to 10km at the Dutch national scale.
- In inversions based on biosphere model parameter optimizations the non-linear model parameters are particularly difficult to constrain. To use them correctly, good priors and a small uncertainty are required, as well as full non-linear model propagation of the solution (rather than a linearized one).
- In contrast, estimates of bias scaling factors on photosynthesis and respiration are more linear, depend less on model structure, and have more freedom to use the diurnal cycle information in the data.
- Still, biosphere model parameter optimisation may be the more promising way forward, because of its ability to ingest other types of observations than atmospheric  $\text{CO}_2$  only (e.g. flux observations to constrain night time respiration, satellite observations to constrain LAI and fPAR, or forest inventory data to estimate carbon stocks).

## Current status emission verification

There are large discrepancies for some greenhouse gases between global “bottom-up” emissions inventories and “top-down” global emissions as determined from atmospheric measurements. (e.g. Weiss and Prinn, 2010 exemplified this for  $\text{CF}_4$ ,  $\text{NF}_3$  and  $\text{SF}_6$ ). Under-reporting of GHG emissions appears to be more common than over-reporting, although both exist. Various factors may tend to bias toward under-reporting, including the price of emissions in carbon-equivalent trading markets and possible unidentified sources.

### European emission verification

Table 3.2 showed the resulting emission estimates per country compared to the prior (JRC estimate) of emission and the value derived by the  $^{222}\text{Rn}$  tracer method. It shows good agreements of  $\text{N}_2\text{O}$  emissions for some countries (NL, B, L) but much higher (50-170%) inverse estimates for others (D, F, and UK. By comparison, (Corazza, Bergamaschi et al. 2011) concluded that their derived total anthropogenic  $\text{N}_2\text{O}$  emissions agree very well with the emissions reported to UNFCCC for most countries: while for UK and Ireland their top-down estimate is about 30% lower than UNFCCC emissions, the agreement is generally better for the other countries: Germany +18%, France -14%



and -11% for the Benelux. They also concluded CH<sub>4</sub> estimated uncertainties of bottom-up and top-down emission estimates are in the same order of magnitude, the uncertainties of the N<sub>2</sub>O top-down estimates are obviously significantly lower than uncertainties of N<sub>2</sub>O bottom-up inventories, demonstrating that inverse modelling can narrow down the overall uncertainties significantly. Similarly, (Bergamaschi, Krol et al. 2010) concluded for CH<sub>4</sub> that inverse estimates agreed well with UNFCCC reported emissions for the UK + Ireland, but hinted at considerably larger emissions for France (+49%), Germany (+71%) and the Benelux (+64%), while in earlier studies (Bergamaschi, Krol et al. 2005) not only French and German emissions seemed underreported but also UK emissions, while Benelux emissions agreed well with inventories. For larger areas the agreement between the two studies was within 6%; the differences for some individual countries were attributed to differences in observational networks between the two studies and improvements in the inversion algorithm and transport models used between the two studies. (Manning, O'Doherty et al. 2011) found for the UK inversion based N<sub>2</sub>O estimated 31% lower than reported to the UNFCCC (2005-2007) but with very similar inter annual trends (1990-2007), while CH<sub>4</sub> emissions were lower by 8% with no consistent trend in the inversions as opposed to the decreasing trend in UNFCCC report. For the same country (Polson, Fowler et al. 2011), based on (late) summertime round-the-island aircraft observations, found high resolution inversion CH<sub>4</sub> estimates 45% higher than bottom up reported and N<sub>2</sub>O emissions even >300% higher. Difference were tentatively attributed to differences in temporal coverage (late summertime being after the peak emission season for N<sub>2</sub>O).

With respect to biogenic emissions (Peters, Krol et al. 2010) present a pixel based estimate of net ecosystem exchange (NEE) of CO<sub>2</sub> in Europe for the years 2001-2007. Over 70 000 atmospheric CO<sub>2</sub> concentration measurements have been used to constrain relatively simple models of terrestrial and oceanic net exchange, while fossil fuel and fire emissions were prescribed. Their method optimises weekly terrestrial sources and sinks for a set of 18 major ecosystems across Europe in which prescribed climate, weather, and surface characteristics introduce finer scale gradients. Their estimates correspond much better with bottom up estimates, the latter derived from data from the flux tower networks and inventory data (Schulze, Luysaert et al. 2009), than a number of previous studies (Janssens, Freibauer et al. 2003).

Reviewing a number of cases (Weiss and Prinn, 2011) argue that is presently already possible to map and quantify regional emissions, and that even with the sparse current network of measurement stations and current inverse-modelling techniques, it is possible to rival the accuracies of regional 'bottom-up' emission estimates for some GHGs. But meeting the verification goals of emissions reduction legislation will require major increases in the density and types of atmospheric observations, as well as expanded inverse-modelling capabilities. They further argue that the cost of this effort would be minor when compared with current investments in carbon-equivalent trading, and would reduce the volatility of that market and increase investment in emissions reduction.

### National scale verification

In the present report first ever attempts have been made to verify bottom-up reported emissions for a small country like the Netherlands. The various results have been collected in table 4.1. The inventory data and their uncertainty in the table are derived from the National Inventory Reports. The uncertainties for the mostly biogenic CH<sub>4</sub> and N<sub>2</sub>O emissions are large and are mostly caused by uncertainties in emission factors rather than by uncertainties in activity data (Olivier, Brandes et al. 2009). The uncertainties at the national scale in fossil fuel originating CO<sub>2</sub> emissions are deemed rather small and slightly dominated by uncertainties in activity data rather than emission factors. The uncertainties in the inversion estimates are similar for all three gases and of the order of 20-30%. Accounting for these uncertainties, the independent atmospheric estimates of Dutch emissions

cannot be qualified as being significantly different from the NIR numbers, yet most numbers as well as the reduction trend suggest emissions may be higher than reported.

Table 4.1.

Emission verification first results for the Netherlands

GHG	Mean emissions Netherlands, 2006-2009				
	inventory	Data inversion <sup>1</sup>	Data inversion <sup>1</sup>	Model inversion <sup>2</sup>	unit
CH <sub>4</sub>	18.3 ± 3.3	22.3 ± 5.6	16.9	23.6	ton km <sup>-2</sup> yr <sup>-1</sup>
N <sub>2</sub> O	12.6 ± 5.7	14.8 ± 3.7	6.71	4.5	10 <sup>-1</sup> ton km <sup>-2</sup> yr <sup>-1</sup>
CO <sub>2</sub> FF	4.5 ± 0.2	4.7 ± 1.6			kton km <sup>-2</sup> yr <sup>-1</sup>
	Emission trend Netherlands, 1992-2000 <sup>3</sup>				
CH <sub>4</sub>	-2.9	-2.2			%yr <sup>-1</sup>
	-34				kton.yr <sup>-1</sup>

1) (van der Laan, Neubert et al. 2009; Van der Laan, Karstens et al. 2010)

2) Vermeulen et al., table 3.2 this report

3) Roemer et al., 2008, pers. comm.

#### Subnational scale/urban centres

With the realisation that 80% of global fossil fuel emissions stem from urban centres around the globe (ref) also reduction ambitions are being expressed by both individual cities as well as urban networks (e.g. the C40Cities, <http://www.c40cities.org/>). In the wake of these realisations also scientific interest to verify emissions at such scales has risen.

From an airborne monitoring campaign over and around Sacramento (CA) (Turnbull, Karion et al. 2011) conclude that their first attempt to estimate urban-scale fossil fuel CO<sub>2</sub> from atmospheric radiocarbon measurements shows that CO<sub>2</sub> FF can be used to verify and improve emission inventories for many poorly known anthropogenic species, separate biospheric CO<sub>2</sub>, and indicates the potential to constrain CO<sub>2</sub> FF emissions if transport uncertainties are reduced. (Djuricin, Pataki et al. 2010) used the <sup>13</sup>C, <sup>18</sup>O, and <sup>14</sup>C tracers to partition between natural gas, gasoline, and aboveground and belowground respiration in the Los Angeles basin. Other recent attempts at atmospheric verification of urban emissions include studies over Indianapolis (Mays, Shepson et al. 2009), Salt Lake City (Pataki, Bowling et al. 2006), Heidelberg (Levin, Hammer et al. 2011) or Debrecen (H) (Molnar, Major et al. 2010) and more studies are on the way (e.g. the NASA/JPL Los Angeles MegaCity CO<sub>2</sub> Pilot Project, Riley et al.; CO<sub>2</sub>-MEGAPARIS project, Xueref-Remy et al.).

Regional emission verification campaigns in more rural areas have been with considerable resolution in e.g. SW France (Dolman, Gerbig et al. 2009). (Lauvaux, Pannekoucke et al. 2009) used concentration measurements from the two tall towers in CERES 2007 campaigns to derive a correction for the fluxes modelled by the ISBA-A-gs coupled to the ARPEGE transport model run at 20km spatial resolution. They found a significant error reduction compared to the prior estimates of land surface fluxes, also on the time evolution of the fluxes, which were both substantially improved by the inversion. The study by (Tolk, Dolman et al. 2011) again pushes the limits of transport modelling and inverse methods to unprecedented resolutions of 10km over the Netherlands (see final sections of chapter 3 this report). Also for CH<sub>4</sub> the present resolution of inversions allow already to draw tentative conclusions on the sub national spatial distribution of emissions. In yet unpublished results Vermeulen et al. (2009, pers. comm.) the posterior maps showed not only a 30% higher national total methane emission compared to the prior, but also a change in spatial distribution.



While in the prior the CH<sub>4</sub> emissions seemed concentrated in the intensive cattle raising areas in the south east of the Netherlands, the posterior shifted the hotspots of emissions more towards the urban areas and wetlands of western Netherlands.

A number of reasons have been identified for differences between (high resolution) bottom-up and inversion estimates:

- Often there is at least some mismatch in the area represented by each estimate: a rather 'vague' footprint vs strict administrative boundaries. When there are considerable emission differences (magnitude and /or trends) in the non-overlapping area the comparisons are flawed (e.g. (Levin, Hammer et al. 2011). When assessing trends one has to realise that these are often less smooth at smaller scales, and that also source fields are much less continuous, but may include strong point sources that need careful representation in prior and transport.
- Similarly there is often considerable mismatch in time frame: data obtained in (aircraft) campaigns are compared vs annual or even longer estimates from inventories.
- Observation network differences are an important source of variations for subsequent estimates for the small region even when done by the same team. This is again a strong reason for long term continuity of the monitoring network
- Though often considered rather precise at national scales the uncertainties in bottom up inventories increases very quickly when zooming in and become as large as those in inversion estimates. Spatial variation is e.g. emission factors that average out at larger scales do not so at higher resolutions.
- Often inventory data are available only in aggregated from at the national scale (as in the NIR). There is a strong need to make inventory info available at the highest possible scale. In some countries steps in this direction are being made (e.g.in the Netherlands they are being made available at the municipality level, as well as on 5km grid)

The most necessary top down ingredients for successful verification systems are thus<sup>1</sup>:

- the ability to make observations at scales similar as that of interest and thus in or close to the areas where emissions/changes are happening (i.e. close to point sources, urban areas, regions, countries)
- a monitoring network that is well harmonised and inter-calibrated, arguably consisting of one or more tall towers within the domain of interest combined a some towers along its borders to allow proper constrains on in/outflow, i.e. the lateral boundaries of the models; co-located monitoring of PBL properties helps interpreting the data
- Further advances in transport modelling both to improve both horizontal transport advection as well as vertical mixing (PBL dynamics). The use of ensembles of models may be used in the meantime to better define transport uncertainties
- Further development of optimal estimation and statistical methods, incorporating all information weighted by precision and accuracy

## Outlook for Verification

The spatial resolution needed for global maps of GHG surface fluxes depends on their final use. For global studies with inversion models, the GEOCS identified the ultimate target spatial resolution as typically 10 km over land and 50 km over the ocean, with a temporal resolution of a week or less (Ciais, Dolman et al. 2010). This can be attained through a coordinated system of integrated global carbon-cycle observations and with significant improvements in data assimilation, atmospheric

<sup>1</sup> For the bottom up elements see the ME1 report.

transport models, and process models of land and ocean carbon cycling. GEOCS short term objective aims at monthly fluxes with spatial resolution of 100 km over land and 500 km over the ocean and is deemed attainable within the next decade (see figure 4.1). However, finer spatial resolutions (sub-hectare to 10 km) are needed for national-level land-use monitoring, reporting situations in the short term for mechanistic studies and verification of compliance with policies, and for detailed mechanistic and validation studies.

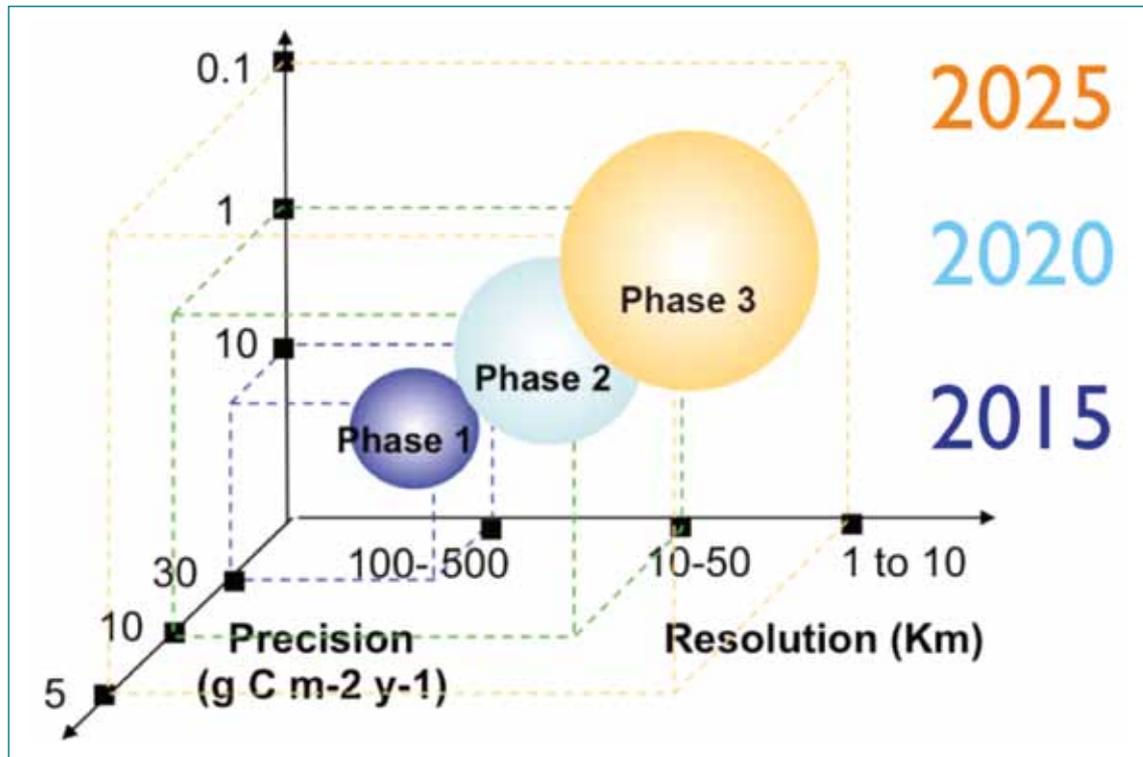


Figure 4.1.

Future evolution of requirements toward finer resolution and precision capabilities for producing global maps of CO<sub>2</sub> and CH<sub>4</sub> surface fluxes according to GEOCS (Ciais, Dolman et al. 2010).

The resolution and precision targets illustrated above and their 'estimated time of arrival' apply on a global scale. For some areas like Europe, N America these might be attained much nearer in the future if the necessary infrastructure can be built and operated with some guaranteed continuity and if the necessary model development can maintain a progression like we have seen over the past few years.

To that end in Europe the ICOS initiative has been launched as one of the vital new European Research Infrastructures under the so-called the European Strategy Forum for Research Infrastructures (ESFRI). ICOS is a European Infrastructure dedicated to high precision monitoring of greenhouse gas balances. It will provide policy makers and scientists with estimates of the fluxes of carbon dioxide, methane, and nitrous oxide, and how these fluxes evolve due to policy measures, climate change, and changes in land use. Besides being motivated by a global perspective (e.g. effects of possible degradation of high-latitude peat soils, and the impact of tropical deforestation), the regional importance is very much centered around verification of national bottom-up emission inventories (as well as estimates of greenhouse gas exchanges with various ecosystem types). Dedicated ICOS-EU centers were recently established to provide services for atmospheric greenhouse gas monitoring (France, Finland, Germany), ecosystem monitoring (Italy), and fossil fuel emissions monitoring (Germany).



On a supra European level ICOS contributes to the implementation of the GEO Carbon Strategy ([http://www.globalcarbonproject.org/global/pdf/GEO\\_CARBNSTRATEGY\\_20101020.pdf](http://www.globalcarbonproject.org/global/pdf/GEO_CARBNSTRATEGY_20101020.pdf)) and the IGACO GHG strategy being implemented by WMO-GAW ([http://www.wmo.int/pages/prog/arep/gaw/gaw\\_home\\_en.html](http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html); see figure below).

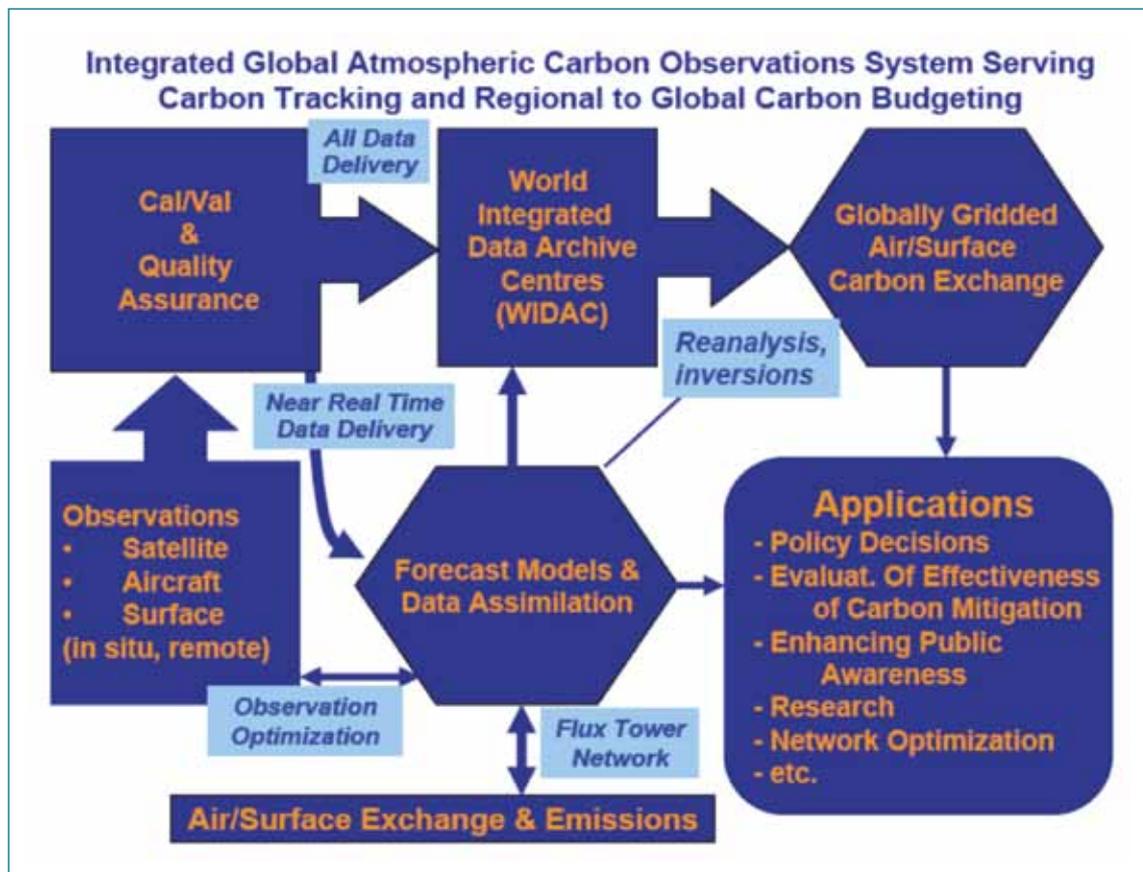


Figure 4.2.

Integrated Global Atmospheric Carbon Observations System serving carbon tracking and regional to global carbon budgeting (source: WMO-GAW)

As demonstrated in the report, in the Netherlands we have built a considerable expertise in the past few years with respect to emission verification both in terms observations and transport modelling and inversion algorithms. The Dutch research network is well imbedded in international efforts as exemplified by participation (often in coordinating roles) of all partners in many international projects, both research (the EU FP6 CarboEurope, FP7 GHGEurope and many smaller projects) and in setting up the infrastructure projects (ICOS, but also e.g. InGOS).

To consolidate this expertise and to better guarantee continuity of observational networks a long term investment and support is needed for infrastructure, beyond the project based funds. To this end also nationally an ICOS-NL initiative has been launched in response to a call targeting large scale research infrastructure. Despite a very high short-list ranking, a first proposal did not get funded. At the time this report is finalised another opportunity for support of research infrastructure has been opened. A new proposal is in preparation in which the Netherlands will contribute an infrastructure for the quantitative and objective interpretation of ICOS-EU observations. The partnership proposes, in addition of a state of art monitoring network, to establish a new ICOS-NL “Carbon Data Portal” to provide the required expertise on high-performance computing, mathematical algorithms, ecosystem-agriculture-climate modelling and model-data fusion techniques. Expertise, research

capacity, as well as policy and science relevant greenhouse balance products will be the core services to Europe. On-going cutting-edge Dutch research in these fields will support the tasks of the centre, and together they open up the perspective of a successful operational GHG emission verification service in the not too far future.

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## Climate changes Spatial Planning

Climate change is one of the major environmental issues of this century. The Netherlands are expected to face climate change impacts on all land- and water related sectors. Therefore water management and spatial planning have to take climate change into account. The research programme 'Climate changes Spatial Planning', that ran from 2004 to 2011, aimed to create applied knowledge to support society to take the right decisions and measures to reduce the adverse impacts of climate change. It focused on enhancing joint learning between scientists and practitioners in the fields of spatial planning, nature, agriculture, and water- and flood risk management. Under the programme five themes were developed: climate scenarios; mitigation; adaptation; integration and communication. Of all scientific research projects synthesis reports were produced. This report is part of the Mitigation series.

## Mitigation

The primary causes for rising concentration of greenhouse gases (GHG) in the atmosphere are fossil fuel combustion, land use and land use change (deforestation). Yet our understanding of interactions between land use (change) and climate is still uncertain. Climate changes Spatial Planning contributed to the development of a system that allows both the best possible 'bottom-up' estimate of the GHG balance in the Netherlands, as well as independent verification 'top-down'. This system supports better management, i.e. reductions of GHG emissions in the land use sector. In this context it addressed a.o. the possibilities and spatial implications of second generation biomass production.

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