ACCENT-BIAFLUX workshop 2005. Trace gas and aerosol flux measurement techniques. Abstract book

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Trace gas and aerosol flux measurement techniques

ABSTRACT BOOK

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The ACCENT-BIAFLUX Workshop 2005, Risoe National Laboratory have 21 oral presentations and 26 poster presentations. The Abstracts are presented in this report.

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Contents

Preface

The Workshop: Trace gas and aerosol flux measurement techniques is the second meeting within the Biosphere Atmosphere Exchange of Pollutants (BIAFLUX) group in the EU-Network project Atmospheric Composition Change (ACCENT) and is held at Risoe National Laboratory, Roskilde, Denmark. The goal of the workshop is to obtain an overview of techniques for measurements of gas and aerosol fluxes and to gather the knowledge of uncertainties in flux measurements and calculations. The workshop is funded by ACCENT.

1 Session: Measurements

1.1 Trace Gas Fluxes from a Micrometeorological Perspective

Dr Steve Oncley
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This talk will review most of the micrometeorological methods commonly (or should be!) used to measure trace gas fluxes. The eddy-correlation/accumulation method is presented, noting frequency response and fetch requirements, averaging times, problems with sampling through tubes (closed-path analyzers), choice of coordinate system, etc. Next are a variety of methods that assume scalar similarity and rely on a calculation of the Modified Bowen Ratio. These methods include flux-profile relations, eddy-dissipation, relaxed eddy-accumulation, and bandpass covariance. Again, the requirements and limitations of these methods -- including the types of assumptions needed are reviewed. Finally, the box-budget method is presented to measure fluxes in difficult situations, such as over complex terrain.

1.2 Development of a Disjunct Eddy Covariance system for VOC fluxes by and PTR-MS

Dr Brian Davison and Ben Langford*
Lancaster University, Environmental Science Department, UK

While continuous eddy covariance may be the best technique for the measurement of fluxes of many parameters few detectors are capable of monitoring at <1Hz. To overcome this Relaxed Eddy Accumulation (REA) was developed by which up drafts and down drafts recorded from the sonic anemometer are sampled onto separate channels with the preconcentrated sample analysed at a later date. This requires a rapid response switching valve to apportion the sample to the correct channel. However, this indirect depends on empirical parameterisations. Also the need for time-integrated sampling means that short-term variations in flux rate are not observable.

An alternative to this is Disjunct Eddy Covariance (DEC). This technique approximates EC by limiting analysis to a sub-sample of eddies, thereby keeping the analysis time for each individual sample short (e.g. < 1 min). A fast grab sample is stored in an evacuated reservoir and analysed off line while the second channel is evacuated ready for sampling. Analysis of VOC's is by an Ionicon PTR-MS instrument. This fast response mass spectrometer allows analysis of several ions with a total analysis time of <1minute.

Virtual disjunct eddy covariance (vDEC) dispenses with the sampling reservoirs and air is sampled and analysed continuously using the fast response of the PTR-MS. The timing of sample air from the intake/sonic, to the detection is critical and to minimise complete analysis time (1sec) a short dwell time is required on each ion monitored. This latter factor increases the signal to noise ratio and makes the vDEC less suitable for species with a low flux rate.

We will discuss some of the important components necessary for development of a DEC system.

1.3 Cost-effective micrometeorological system for measuring landfill gas emissions

Tuomas Laurila[1], Juha-Pekka Tuovinen[1], Annalea Lohila[1], Mika Aurela[1], Tea Thum[1],Mari Pihlatie[2], Janne Rinne[2] and Timo Vesala[2]
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Landfills are an important source of methane to the atmosphere that should be reported to the authorities making inventories of the greenhouse gas emissions. For the maintenance of landfill gas recovery systems and top sealing structures, continuous emission data of methane and carbon dioxide would be of primary importance. Chamber techniques have been used to measure the gaseous emissions, but they are laborious, and it is difficult to obtain representative emission estimates due to the high spatial variability of fluxes. Spatially integrated landfill emissions have been measured in tracer experiments, but complex landscapes may limit the use of this method. Moreover, it is suitable only for methane, and it is difficult to acquire continuous time-series. We have used micrometeorological methods to measure landfill gas emissions to the atmosphere on a continuous basis. In this presentation, we describe an eddy-covariance measuring system for landfill gas fluxes and present some results from a field experiment.

We have developed a micrometeorological system for the CH_4 and CO_2 fluxes and evaporation. The instrumentation includes a three-dimensional sonic anemometer and a LI-Cor LI-7500 sensor for CO_2 and H_2O concentrations. For the measurement of CH_4 concentration, air was drawn at a high flow rate to a Flame Ionization Detector (FID). Fluxes measured by the FID-based system were verified by those observed using a Tunable Diode Laser detector, the two methods showing very similar results. The experimental site was located in the Ämmässuo landfill in the Helsinki metropolitan area, which is the largest landfill in Finland. The landfill is in active use and there are also some recent deposit areas close to our measurement site. A gas recovery system is in operation in all parts of the landfill. The CO_2 and H_2O flux measurements began in February, 2003 and the CH_4 flux measurements in June 2003 continuing until December 2003.

Evaporation measurements are needed for understanding the hydrology of the landfill body and the formation of leachate water. We observed average noontime evaporation rates between 30 and 60 mg H_2O $m^{-2}s^{-1}$ during the summer months. The total monthly evaporation was highest in May, about 60 mm, but lower in June and July when precipitation was limited. The CO_2 and CH_4 fluxes were highly dependent on wind direction, as was their ratio. This reflects the fact that these gases originate in the landfill body and that their emissions depend on both diffusion through the spatially variable surface soil layer and the direct routes such as cracks and wells. The highest rates were

typically observed in the directions in which there are well structures and recent waste deposits. The average fluxes during July–December were about 2 and 0.5 mg m^{-2} s^{-1} for CO_2 and CH_4, respectively. A comparison of the micrometeorological fluxes to chamber measurements showed that they were of the same order of magnitude, but the spatial variability of the chamber measurements was very high.

1.4 Particle fluxes above forests

Dr. Sara C. Pryor
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Atmospheric resistance to atmosphere-surface transfer is typically low over forests due to the high roughness and well developed turbulence. Hence, the flux may be significant even when the vertical concentration gradients are small and this flux may substantially alter the in situ particle spectrum and concentration. Although several process-level models have been developed to simulate dry deposition of particles to vegetated surfaces and the degree of specificity of the models is variable, the model most frequently applied to forest canopies is that of Slinn (1982). While there has recently been validation of this model for particles with diameters (d_p) ≤ 0.1-0.2 μm, there are substantial and systematic discrepancies between process-level models of particle dry deposition and available measurements, particularly for $d_p \approx 0.2 – 1$ μm and forested surfaces. Models such as those proposed by Slinn (1982) typically predict particle v_d in this size range of the order of 0.1 mm s^{-1} while measurements imply values that are an order of magnitude higher. A key assumption in application of this model is that, with the exception of deliquescent behavior, the particle size distribution is invariant during the dry deposition process, and hence the deposition calculation can be effectively decoupled from the rest of the particle dynamics. I will present model experiments designed to test the hypothesis that violations of the constant flux layer assumption can be invoked to explain part of the discrepancy between the models and measurements (Pryor and Binkowski, 2004). I will also present data from a campaign conducted to examine evidence of flux divergence due to particle dynamics.

References:

Pryor S.C. and Binkowski F.S. (2004): An analysis of the time scales associated with aerosol processes during dry deposition. *Aerosol Science and Technology* **38:** 1091-1098.

Slinn W. (1982): Predictions for particle deposition to vegetative canopies. *Atmospheric Environment* **16**: 1785-1794.

1.5 Spectral methods for estimation of fluxes of scalars

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Fluxes of gases and particles can be measured directly by the eddy correlation technique if fast response instruments are available. However eddy correlation technique, used at moving platforms like ships or where flow distortion is pronounced can give erroneous results. Under such conditions spectral methods are preferable because here it is possible to filter out the effect from the distortion. Also fluxes of scalars measured using instruments with slower sampling time (< 1Hz) like NO_2, SO_2 and O_3 can be estimated from idealized spectral shapes assuming that turbulence transport is the main transport process.
Here we present fluxes of particles, CO_2 and water vapor over forest area estimated from co-spectra and power spectra analysis using both high frequency sampling (10-20Hz) and low frequency sampling (1Hz).

1.6 Estimation of emission fluxes from a horizontal flux budget method, exemplified with determination of pesticide volatilization

N. O. Jensen, Helle Vibeke Andersen[*]
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The paper describes an experimental set-up designed to measure the evaporation of different pesticides after application under full-scale field conditions. Two campaigns were conducted on the same location: one on bare soil during fall and one in spring on a barley crop.The pesticides were sprayed around the circumference of a circle and mean air concentration samples (averaging time from 1 hr to 6 hr, increasing gradually during the week long sampling periods) were taken in the centre of the circle at different heights up to 9 m above ground. The wind speed, friction velocity and sensible heat flux were measured by a sonic anemometer (Gill) and evaporation (latent heat flux) was determined from an infrared absorption sensor (Ophir). Auxiliary measurements of e.g. mean air temperature and soil humidity (by TDR) were also made. The advantage of this design is that the sprayed area is very small (short application time) so that the initially large fluxes can be followed right from the beginning. In contrast this would not be possible by traditional methods such as the classical profile method, eddy correlation, or REA, since the sprayed area would have to be very large, in order to obtain horizontal homogeneous conditions, and consequently the application time would be too large compared to the time scale of the initial course of the volatilization. A further advantage is that the fetch from the sprayed area to the sampling position ensures that the near ground concentration gradient is reduced in steepness (reducing error in the necessary extrapolation) and that the circular form makes it possible to sample under invariant conditions even during day to day changes in wind direction. Details on the determination of the dimensions and geometry of the experiment are given in the paper. A method to increase the confidence of the flux calculations is also described. It utilizes that several substances was applied simultaneously and that a certain parameter in the fitted concentration profiles depends on turbulence only and thus have the same value for all substances.

1.7 (P1) Eddy Covariance Particle Flux and Size Distribution Measurements above Norway Spruce

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In July / August 2001 and 2002, measurements were carried out in the "Fichtelgebirge" mountain range (50°09' N, 11°52' E) within the framework of the joint project BEWA2000 of the German atmospheric research programme AFO2000 to study turbulent particle fluxes as well as particle formation and growth involving reactive organic compounds. An eddy covariance system was set up above a "Norway spruce" forest combining a Young 81000 ultrasonic anemometer and two condensational particle counters (TSI UCPC3025 and TSI CPC3760A) to measure turbulent particle exchange. The time evolution of the submicron particle size distribution was measured above the stand with a twin differential mobility particle sizer (TDMPS). In addition, particle number concentrations were measured within the forest stand 4 m above the ground using a TSI UCPC3025 particle counter.

Particle concentrations within and above the forest differed considerably. The number concentration within the forest ranged from 60 % to 100 % of the concentration above the forest and exhibited a larger amplitude in the diurnal pattern compared to the measurements above the forest. From these findings we conclude that not only particle concentrations but also particle size distributions within and above the forest are different from each other. This has to be taken into account when performing a comparative analysis of particle flux and size distribution measurements.

On most days, a diurnal pattern of the turbulent particle number fluxes with small or no fluxes during night-time and strong fluxes around noon was found. In general, deposition fluxes dominated over emission. However, brief particle emission events had a distinct impact on the size distribution of the particle population. Particle formation events were identified on several days, exhibiting a characteristic "banana-shaped" evolution of the particle size distribution. The newly formed particles showed a remarkably constant growth behavior with typical diameter growth rates ranging from 2 to 6 nm h^{-1}. Condensation of sulfuric acid is not sufficient to maintain these growth rates. A more detailed analysis of the growth dynamics revealed the importance of the condensation of oxidation products of biogenic volatile organic compounds (BVOC) for the observed particle growth.

During particle formation events, the strongest particle fluxes were observed, and also, particle deposition dominated over emission. Given the observed particle growth rates, we conclude from these facts that particle formation takes place effectively above the forest stand.

1.8 (P2) Eddy covariance flux measurements of NO and NO_2/NO_y over a mixed deciduous forest in a moderately polluted environment – description of measurement system, evaluation algorithms and quality assessment

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In 2002 and 2003, two large-scale field experiments were conducted over a deciduous forest at Juelich (Germany) within the framework of AFO2000-ECHO (Emission and CHemical Transformation of biogenic volatile Organic compounds). Fluxes of momentum, sensible heat, H_2O, and CO_2, as well as of reactive trace gases O_3, NO and NO_2/NO_y have been measured by eddy covariance (EC) techniques at 30 m (above ground) on the ECHO-"westtower".
NO and NO_2/NO_y-measurements were simultaneously performed using a fast, highly sensitive 2 channel chemiluminescence-based NO-analyzer (CLD 790 SR, EcoPhysics). Alternatively, we used either (in 2002) a NO_y-converter (in-house development), or (in 2003) a photolysis converter (PLC 81800, ThermoOriel).
We will present the instrumentation, the data-acquisition, and the resultant cospectral behaviour of the system. Next, we will discuss the applied evaluation and correction algorithms in detail.

1.9 (P3) Comparison of O_3 fluxes measured with the eddy covariance and the gradient methods above a maize field.

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Between June and September 2002, the fluxes of carbon dioxide (CO_2) and ozone (O_3), and nitrogen oxides (NO and NO_2) were measured above a growing maize crop, together with the components of the energy balance - sensible (H) , ground (G) and latent heat fluxes (H), net radiation (R_n) – , the meteorology, and the canopy structure. All the fluxes, except the NO_x, were measured simultaneously with the gradient and the covariance technique. The gradient mast consisted in 9 levels extending from 0.3 to 10 m above ground, where CO_2 and H_2O was measured with a Licor Li6262, and O_3 and NO_x was measured with two separate analysers (2 Environnement SA 41M for O_3 and 2 ThermoEnvironnement 42C for NO_x), each screening the 5 uppermost and the 5 lowermost levels, with a common level allowing for cross calibration. Wind speed and temperature was also measured, and the fluxes were determined using 3 to 4 levels above the canopy. The covariance mast consisted in a Gill R3-50, a Licor Li7500 and a fast O_3 chemiluminescent based close-path sensor from ATDD V2.0 with solid-plates “targets” coated with Coumarin. Two identical covariance masts were compared during 15 days, to determine the lifetime of a “target”. In this study, the momentum, energy, CO_2, and O_3 fluxes estimated with the gradient method and the two eddy-covariance masts are compared, with a special focus on O_3.

1.10 (P4) Continuous measurements of O_3 fluxes by covariance over forest and arable cropland in France

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Terrestrial ecosystems are significant sources of atmospheric gases such as reactive nitrogen (NH_3, NO and N_2O). Conversely, they can be a sink for NH_3, NO_x, and O_3, which can lead to several adverse effects (acidification and eutrophication, direct effects). But deposition also contributes to decreasing their lifetime in the atmosphere, and is therefore an efficient "depollution process". In this context, continuous measurements of O_3 fluxes as well as other compounds have been set up at two sites in France.

The fluxes of O_3, CO_2/H_2O are measured continuously with the eddy covariance method over the "LeBray" pine forest near Bordeaux since 2003, and over a cropland field near Paris since mid-2004. The Bray forest site is 35 year old plantation of maritime pine (*Pinus pinaster Ait*.) with an understorey of graminae. It is currently a CarboEurope site and has been part of the Euroflux network since 1996. The trees are distributed in parallel rows along the NE-SW axis. The inter row distance is 4m and the stand density is about 500 trees ha-1. The mean tree height is 20 m and the LAI around 2.6. The site is flat and the fetch is over 600 m in the prevailing wind direction. The climate is oceanic with 900 mm of rainfall per year. Instruments are installed at 43 m.

The cropland site is a CarboEurope site, with a rotation of mustard, maize, wheat and barley, with a nitrogen supply of 120 kg ha^{-1} y^{-1}, as mineral fertiliser, and an additional 100 kg ha^{-1} every three years as cattle slurry. The climate is continental-oceanic, with 600 mm of rainfall per year, the soil is a luvisol with 60% of silt, and 30% of clay. Instruments are installed at 2.5 to 4.5 m.

At both sites, the eddy-covariance equipment set includes a Gill RGA3-50 or a Gill R2 sonic anemometer, a H_20 and CO_2 Licor 7500 analyser, and a fast O_3 chemiluminescent based close-path sensor (ATDD V2.0) with solid-plates "targets" coated with Coumarin. Additional absolute measurements are performed at both sites. Automatic chambers measuring N_2O and NO emission will be installed in 2005 in Grignon, as well as NH_3 gradient measurements. Initial results from these two sites will be presented.

1.11 (P5) Use and validation of a Relaxed Eddy Accumulation (REA) System to measure water vapor, carbon dioxide and nitrogen oxides fluxes over a maize field.

Aurore Brut
CNRM/ Météo-France

Increasing interest in dry deposition of air pollutant and trace gas flux determination has widely encouraged the development of the Relaxed Eddy Accumulation (or conditional sampling) technique. This technique remains very attractive since many trace gas constituents cannot be measured fast enough to allow the application of the eddy correlation method. This technique relies on sampling turbulent air according to the sign of the vertical wind fluctuations (w') in order to collect, at a constant flow rate, updraft air in one reservoir and downdraft air in another. At the end of the sampling period, the mean concentration in each container is measured and the flux is proportional to the difference of both concentrations.
A Relaxed Eddy Accumulation system, developed at CNRM, was implemented above a maize field near Meyrargues during the ESCOMPTE experiment. It operated continuously surface flux measurements of water vapor, carbon dioxide and nitrogen oxides for almost 4 weeks. In addition, two other Eddy Correlation systems were settled close to the REA device : the first one implemented by INRA measured both humidity and CO_2 fluxes with a Li6262 sensor whereas the second one, operated by CNRM measured water vapor fluxes with a fast hygrometer.
Comparisons of humidity and carbon dioxide fluxes, measured by the various systems are presented. The good agreement (high correlations, small biases) between fluxes tends to validate the conditional criterion of the REA selection system. The NO_x fluxes represents an original dataset for which, a further study allowed the determination of parameterizations according to meteorological conditions (Fotiadi A., 2003).

1.12 (P6) The dependence of the β-factor of REA system with dynamic deadband on atmospheric conditions

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The most direct method for measuring fluxes is the eddy covariance (EC) method which requires fast-response sensors. An alternate method, the relaxed eddy accumulation (REA) requires that air be sampled at a constant rate and placed in one container during the upward wind velocity (w) and in another during downward wind velocity. Since this method is based on accumulated concentrations, slow response sensors are sufficient.
To increase the concentration (c) difference and to reduce the wear of sampling valves, a dead-band around $w = 0$, in which air is not sampled, was introduced into the system. The constancy of the sample flow rate introduced an empirical constant, β, into the flux equation

$$F_c = \beta\sigma_w(\overline{c^{\uparrow}} - \overline{c^{\downarrow}}), \quad (1)$$

where σ_w is the standard deviation of w. The value of β depends on the dead-band width, which in our system is proportional to the running mean of σ_w (dynamic dead-band).
In this study the dependence of the value β -parameter of the REA technique with dynamic dead-band on atmospheric conditions and its uncertainty are studied systematically. This is conducted by simulations using wind velocity and carbon dioxide measurements at the rate of 10 Hz. The tower for the measurements is located in a Scots pine (*Pinus sylvestris* L.) forest with dominant tree height of 14 m at SMEAR II station, Hyytiälä, Southern Finland. The measurement height was approximately at 23 m from the ground, about 10 m above the forest canopy.
We calculated fluxes according to REA and EC principles for each 30 minutes averaging period. EC flux was given by the covariance between the w and c ($w'c'$). As in our REA software, we calculated the dynamic dead-band value, $\pm 0.5\sigma_w$, using the 5 minutes running mean. Based on the dead-band and w, we determined the REA valve position. Depending on that, we updated the concentration and the number of measurement values of up, down or dead channels. After 30 minutes period we calculated the REA flux.
After that we estimated the coefficient β for each averaging period of turbulent fluxes from

$$\beta = \frac{\overline{w'c'}}{\sigma_w(\overline{c^{\uparrow}} - \overline{c^{\downarrow}})}. \quad (2)$$

To see the effect of dead-band width, we repeated simulations using the dead-band width proportional to $(0.2\text{-}0.8)\sigma_w$.

(P7) Field intercomparison of disjunct and conventional eddy covariance systems

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The disjunct eddy covariance and disjunct eddy accumulation methods have recently been utilized to measure fluxes of such trace gases for which fast on-line analyzers do not exist. A field intercomparison experiment of a disjunct eddy covariance (DEC) and conventional eddy covariance (EC) systems was conducted over short grass field. A DEC system constructed for the experiment consisted of two intermediate storage reservoirs used to grab near-instantaneous air samples and an infrared gas analyzer to analyze water vapor concentrations in the reservoirs. The samples were taken in 0.1 seconds with 15 second interval leading to 120 samples per half an hour averaging period. The EC system measured water vapor flux by an acoustic anemometer and a krypton hygrometer. During two days the water vapor fluxes measured by the DEC system were within the estimated uncertainty from the flux measured by the EC system, with good correlation (r^2=0.94). There was a slight systematic overestimation of the fluxes measured by the DEC system during the day and underestimation during the night leading to relation $F_{DEC}=1.15*F_{EC}-9.69$ W m^{-2}. As the bias doesn't appear in the simulated DEC measurements it is likely to be due to instrumental rather than methodological problems.

1.13 (P8) Ozone uptake by an evergreen Mediterranean forest (Quercus Ilex) obtained by micrometeorological flux measurements in Italy.

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Ozone, water and energy fluxes have been measured over a Mediterranean evergreen forest in Central Italy from August to October 2003 with the eddy-correlation technique in order to evaluate the amount of ozone taken up by plants in dry summer and in mild autumn conditions. The stomatal ozone fluxes has been calculated using the analogy with water vapour fluxes inside the stomata, which are easily measurable. The total ozone dose was obtained by integrating the stomatal fluxes over time.
Stomatal flux resulted a minor part (31.5%) of the total ozone flux over the forest ecosystem. The main part of ozone deposition follows non-stomatal pathways. Chemical sink seems to play a relevant role in the morning non-stomatal deposition. Stomatal uptake is enhanced by water availability but, on the average, it does not exceed the 34.4% of the total ozone flux.
A comparison between the cumulated stomatal ozone fluxes and the currently used AOT40 exposure index highlighted important distortions introduced by this index. AOT40, which do not take into account plant physiology, lead to substantial overestimation of ozone risk, particularly when water supply is limited, as occurs frequently in Southern European and Mediterranean areas.

1.14 (P9) Relationship between evapotranspiration and ozone fluxes at agricultural ecosystem level in *Allium Cepa* L.

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Ozone risk assessment for crops requires the determination of the ozone stomatal fluxes F_{ST} instead of the simple ozone exposure (AOT40).
Ozone and water fluxes were measured continuously at agricultural ecosystem level for a relevant part of the growing season of an Allium cepa crop field in 2003. Stomatal ozone fluxes were deducted from evapotranspiration by inverting the Penman Monteith equation.
The cumulative ozone stomatal flux of the crop, i.e. the toxicologically effective ozone dose, resulted 13.674 mmol/m^2 versus an ozone exposure of 5165 ppb·h for the same period.
The bioavailable ozone fraction (Fst/Ftot) was the 43.4% of the total ozone entering the agricultural ecosystem, a result that is very similar on how previously observed in other crops (wheat, barley, soybean)
A significant amount of the ozone stomatal fluxes occurred at ozone concentrations below 40 ppb. This fact rise some questions about the opportunity to adopt any ozone accumulation threshold.
As this crop needs to be constantly irrigated, the ozone stomatal resistance resulted strongly influenced by the irrigations and was only slightly dependent by the other environmental factors even if these latter occurred at severe levels.
As a consequence, in this case the ozone stomatal fluxes resulted relatively coupled to the ozone exposure expressed as AOT40.

1.15 (P10) A flexible automated dynamic chamber system for measurement of reactive trace gas fluxes

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To measure reactive trace gas exchange between low vegetation and atmosphere with respect to different plant composition and management practices, quasi-continuous and parallel flux measurements on multiple experimental plots are necessary. For this purpose, we developed an automated and mobile dynamic chamber system, which guarantees minimum disturbance of the environmental and chemical conditions of the plants and soil enclosed. The system consists of a maximum of six chambers, which are measured sequentially. The chambers have a modular design and are easy to mount. Therefore, they can be used for rapid screening of different plots or easily be removed during management procedures. They have a circular cross section (diameter: 0.4 m) and a flexible volume (40 - 90 L). The chamber walls consist of highly transparent (for PAR and photolysis frequencies) and chemically inert PFA film to minimize wall deposition effects of reactive species (e.g. VOC). The controllable lids are closed during sampling periods (8 - 20 min) only, in order to maintain ambient conditions as frequently as possible. High ambient air flow rates (60 L min-1) assure rapid exchange of chamber air. Integrated flexible control and data acquisition units provide continuous monitoring of important parameters, e.g. in-chamber air temperature and relative humidity, soil temperature, and soil moisture for each individual chamber. The presentation will give a detailed description of the setup of the operational chamber system. We will present examples of flux measurements for several trace gases from various field experiments. Comparisons to other flux measurement techniques, e.g. eddy covariance, will prove the applicability of the method.

1.16 (P11) Laboratory measurements of NO_x and N_2O emissions from soil cores

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Within the NOFRETETE (Nitrogen Oxides Emissions from European Forest Ecosystems) project, a new laboratory system for NO_x (dynamic) and N_2O (static) measurements from intact soil cores was constructed. NO_x emissions from up to 12 soil cores are measured fully automatically using adapted Kilner jars as flow-through chambers. One empty jar serves as reverence chamber. The flow-through is constant at 1.5 l min^{-1} and is generated from the NO_x-analyser pump. To switch from NO_x to N_2O measurement the chamber ports are closed and air is sampled with a syringe via removable septa. N_2O is measured with gas chromatography and fluxes are calculated from linear concentration increase over incubation time.
The system is constructed primarily to investigate soil temperature and soil moisture effects on nitrogen oxides emissions. Therefore the chambers are situated in an incubator/refrigerator with temperature control (0-45°C). Soil moisture can be adjusted using pressure plates or by specific soil core weights for different water contents. Soil moisture is kept constant through periodic rewetting.
We will present (1) a detailed description of the measuring system, (2) results from the NOFRTETE project, where temperature and moisture effects on soils from Finland, Germany, Austria and Italy have been investigated in the laboratory, and (3) compare the laboratory results with NOFRTETE field sites investigations.

1.17 (P12) Nitrogen oxides emission from two beech forests subjected to different nitrogen loads

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We measured nitrogen oxides (N_2O and NO_x) and carbon dioxide (CO_2) emissions from two beech forest soils near Vienna, Austria, which were exposed to different loads of nitrogen input from the atmosphere. The site Schottenwald (SW) was receiving 22.9 kg and Klausenleopoldsdorf (KL) 13.4 kg nitrogen via wet and dry deposition. Nitrous oxide and CO_2 emissions were measured with manually operated chambers biweekly (SW) or monthly (KL). Additionally, daily N_2O measurements were carried out with an automatic gas sampling system. NO_x emissions from soil were detected hourly with an automatic dynamic chamber system. We used the autoregression procedure (time-series analysis) to estimate the relationship between nitrogen oxide emissions and several climate, soil chemistry and N-deposition data. Changes in soil moisture and soil temperature had the most profound effect on CO_2 and on nitrogen oxide emissions and could explain up to 95% of the temporal variations of gas emissions. The spatial and temporal variability of nitrogen oxide fluxes was high. Event emissions after rain or during freezing and thawing cycles made up a high proportion (for NO 50%) of overall nitrogen oxides emissions. In the two investigation years, annual gaseous N losses ranged from 0.65 to 0.77 kg N_2O-N ha^{-1} y^{-1} and from 0.18 to 0.67 kg NO-N ha^{-1} y^{-1} in SW. In KL significantly lower annual N_2O emissions were determined (0.52 kg N_2O-N kg ha^{-1} y^{-1}). In KL a measuring campaign, revealed a NO-loss of 0.02 kg, whereas in the same time significantly more NO was emitted in SW (0.32 kg NO-N ha^{-1} y^{-1}). Higher nitrogen oxides, especially NO emissions from the high nitrogen input site (SW) indicate that atmospheric deposition had a strong impact on losses of gaseous nitrogen from our forest soils. At KL stronger correlations were detected between N-emission and nitrogen deposition, indicating that low N-input sites are more responsive to increased nitrogen input than N-saturated sites.

2 Session: Low-Cost Techniques

2.1 Long-term measurements of reactive trace gas fluxes by time average gradient

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2.2 Development of a low cost system for measuring deposition fluxes on the field scale by micrometeorology

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The main goal of the Time Averaged Gradient system (from now on referred to as TAG) is to provide direct long-term average (1 to 4 weeks) flux measurements for a range of trace gas species, between atmosphere and terrestrial surfaces.
Over daily periods, atmospheric conditions can range from high stability, where the vertical gradients of ambient concentration are enhanced due to very small diffusivity, to highly unstable conditions, in which concentration gradients are small due to the intense turbulent activity of the surface layer.
By sampling continuously over a long-term period, the large vertical gradients generated by high stability would lead to an over-estimate of the actual flux. It is necessary, in developing the TAG system, to develop techniques which avoid the bias due to these processes. To overcome this problem, the TAG system operates conditionally, sampling the micrometeorological variables within a carefully defined range of stability.
The development of such system is described, and an analysis of the outcome of its conditional protocol is presented, showing a comparison between the TAG system and a continuous flux gradient system that were operating on the same field site.
The reliability of the TAG conditional technique to detect fluxes of trace gases between atmosphere and vegetation is discussed.

2.3 COTAG nitrogen flux measurements in Norway supported by the EMEP deposition module

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This paper presents nitrogen flux measurements carried out by the Conditional Time Average Gradient (COTAG) method at an alpine site (700 masl) in Storgama in Telemark, Norway. As part of the five year multi discipline project "Effect of Climate and Land Use Change of N&C fluxes; Air Land Freshwater Marine Links" (CLUE - http://www.nlh.no/ipm/forskning/clue/), two COTAG systems were established in June 2004. Filter packs rather than denuders were used to sample the main compounds in the air and more powerful pumps were used in contrast to the traditional COTAG setup. The averaging period was two weeks. In addition to the alpine site at Storgama, a system was installed in the nearby village Treungen. However, at this latter site, meteorological conditions were favourable (near neutral conditions, i.e. Richardson number close to zero) for sampling only for a small fraction of the time, and it is uncertain whether the measurements are representative. Here, we present results from June to December 2004, i.e. through three seasons, with a thin snow cover on the ground in the last sampling periods. Measurements will continue after spring thaw in 2005. The system worked well in the entire period, and sampling frequency was typically more than 50%. The results show large variability in fluxes and deposition velocities. The EMEP deposition module was used both on continuously sampled meteorological data and on data from the periods when the COTAG was sampling. As expected it shows much less variability than the measurements, but gives deposition velocities of the same order of magnitude as the COTAG in both runs.

2.4 (P13) Scanning biogenic NO emissions from soils: a reasonably simple, automated laboratory soil chamber system for realistic ambient conditions

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A reasonably simple, automated laboratory (dynamic) chamber system is described, which allows rather fast scanning of biogenic NO emissions from soil samples. The influence of (a) realistic ambient nitric oxide (NO) mixing ratios, (b) soil moisture, (c) soil temperature, and (c) soil nutrient availability on NO production and consumption, as well as on soil compensation mixing ratios of NO can also be quantified with that system. Four measurement chambers with soil samples as well as one reference (control) chamber made of polyacrylic glass are flushed continuously with air at a rate of 2.5 L min-1. The chambers are placed in a thermostat cabinet to control the soil temperature (0-30°C). Inverted gas drying tubes are introduced into the system to maintain soil moisture at prescribed, but constant levels (0% to field capacity). A gas dilution system provides various NO mixing ratios (0-200 ppbv), and a commercial chemiluminescence NO analyzer is used to measure the NO mixing ratio in the headspace of individual chambers. NO emission (and NO uptake fluxes, respectively) are estimated from soil production and consumption rates with the help of a modified Galbally & Johansson algorithm (Galbally & Johansson,1989). We will present (a) a detailed description of the system, (b) examples of derived results from South African, Brazilian, and Mid European ecosystems, and
(c) validation of the laboratory estimates by corresponding results from field dynamic chamber measurements.

3 Session: Corrections to flux measurements

3.1 The Eddy Covariance flux equation

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The design of any physical instrument or measurement system must be based on fundamental physical principles. Eddy covariance technology and the mass and energy fluxes derived from this method are no exceptions. This presentation reviews and discusses the (3-dimensional) mass conservation equation supporting the measurement of eddy covariance fluxes. For the measurement of (vertical) trace gas fluxes by micrometeorological methods located above an active surface, this conservation equation includes two crucial components: one associated with the in situ measurement of the trace gas itself, the other associated with the measurement of atmospheric density effects (the WPL terms). The true surface exchange flux combines these measurements. However, for many trace gases the WPL terms are by far the dominant terms, which in turn introduces greater uncertainty in the final estimate of the true surface flux. Historically, the WPL terms were justified by relating the density effects to a mean vertical velocity. But such an approach makes it difficult to clearly distinguish WPL effects from vertical velocities associated with either true advective motions or coordinate rotation. This confusion is avoided by combining the equations of mass conservation for both the trace gas and dry air. The benefit of this approach is that it removes any ambiguity concerning the WPL effects and each term in the resulting equation can be identified with a physical effect. However, the cost is the introduction of additional terms, the quasi-advective terms, which have not been previously identified or evaluated for their contribution to (vertically integrated) mass balance equation. This presentation includes a discussion of the importance of these additional terms.

3.2 Application of ogive analysis for quality control and spectral correction of eddy covariance fluxes

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One big advantage of the eddy covariance (EC) method compared to other flux measurement techniques is that, beside the average flux value, the high frequency time series contain much more information about the spectral turbulence structure which can be used for quality control purposes and even for correction of the EC flux. With strongly increased computer power and data storage capacity available nowadays it is relatively easy to perform operational spectral analysis of EC data. However, optimized data reduction and evaluation strategies for quantitative spectral information are important.
As part of the operational EC flux calculation for a CarboEurope grassland site, we performed an evaluation of spectral information on a dataset of several months. The main problems for flux measurement at the grassland site are on the one hand a limited fetch that implies potential footprint problems and necessitates a low measurement height of typically 1 m above the grass canopy. This leads to the dominance of small-scales in the turbulence spectra susceptible to high-frequency damping effects due to sensor separation or tube sampling. On the other hand, calm night-time conditions lead to frequent breakdown of turbulence.
For each half-our interval the cumulative cospectra (ogives) for fluxes of momentum, sensible and latent heat, CO_2, and other trace gases were calculated and reduced to 40 points by selection of logarithmically spaced frequencies. The reduced ogives were analysed visually and numerically to give quantitative information about the breakdown or intermittency of turbulence, about disturbance effects on the low frequency end and damping effects on the high frequency end of the spectra. The latter effect was analysed by comparing the normalised ogive of trace gas fluxes to the respective ogive of sensible heat flux. For certain conditions, the experimentally obtained damping factors deviated considerably from the results of the commonly applied theoretical parameterisation after Moore (1986). This is especially important for the CO_2 flux, because it requires a very high accuracy level for the calculation of significant annual carbon budgets.

3.3 Low pass filter correction to turbulence data using a digital recursive filter

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In many long-term flux stations eddy covariance systems are used, where gas intake and gas analysis are separated by several tens of meters. It has been recognised that the gas lining via tubes introduces both a low pass filtering effect and a time lag to the measured signal compared to the true turbulent concentration course. The physical causes are: mass flow through a death volume, mixing by turbulent diffusion along the flow, and adsorption and desorption to and from the wall material and possible deposits on the surfaces. All these effects decrease the absolute covariance between the vertical wind speed and the concentration and have thus to be considered in flux calculations.

Theoretical treatment of the effects is difficult, because of the many unknowns in flow distribution and surface interactions of the gas in the tube systems including funnels, filters, valves, etc.. Also the empirical derivation of a correction turned out difficult, because the corrections overlap and interfere with part of the data post-processing. All published correction procedures have difficulties to cope with the problem of measuring the unbiased turbulence with the help of turbulent air temperature, when absolute sensible heat fluxes are low. For this reason, parameterisation of low pass filtering effects has advantages over performing spectral corrections separately for each single half-hourly turbulence time series.

Using turbulence data from a beech forest and a grassland site, a simple and accurate empirical parameterisation of the low pass filtering effects was developed. The parameterisation is based on a digital recursive filter and considers influence of horizontal wind speed. The resulting correction was compared with parallel measurements using an open path system. Our findings show that the low pass filtering effects differ between the grassland and the forest sites, and, that they change with wind speed, atmospheric stability, duration of tube operation, and tube temperature. Stronger interaction between water vapour and the gas lining system was reflected by smaller low pass filter constants. Consequently, absolute effects were generally larger for water vapour than for carbon dioxide.

The parameterisation was then used to correct long-term flux data. Because the low pass filtering effects were higher at night, the correction increased respiratory carbon dioxide effluxes. Consequently, the annual net carbon dioxide budgets were substantially lower after low pass filtering correction.

Our findings convinced us that this correction is necessary for a more realistic flux estimation, if long tubes or other influences cause low pass filtering in an eddy correlation system.

3.4 Corrections to flux measurements from moving platforms

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The platforms available for flux measurements over the open ocean, whether ships or buoys, are invariably moving. In order to obtain data in high sea states, ships are more reliable as instrument platforms than buoys, but create a much greater obstruction to the mean air flow. Corrections for both motion and air flow distortion are required if mean and fluctuating quantities are to be determined to useful accuracy. We will quantify the likely errors and review the necessary corrections using our own, and other published, research.

3.5 The Importance of Stationarity in Eddy-Correlation WorkMethods of Quantification

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The eddy-correlation technique is widely used to determine surface fluxes of atmospheric minor constituents, both natural and anthropogenic, upward and downward. It makes use of one of the basic assumptions of micrometeorology, which consists in stating that, after having expressed all turbulent variables as sums of their time averages and their fluctuations, the time averages obey the same laws of fluid dynamics as the instantaneous values do. Vertical fluxes are then equalled to the covariances of the fluctuations of the physical variable transferred vertically and of the vertical component of the wind vector, respectively.
The underlying, implicit assumption equalling fluxes with covariances between fluctuations is that turbulence is the driving force originating vertical fluxes, and that there is a decoupling between fast fluctuations generated by turbulence and slow fluctuations due to synoptic processes. This decoupling is observed only when the time series of the turbulent variables show stationary behaviour. Stationarity of the temporal variations (fluctuations) represents thus a necessary condition for obtaining reliable data with the eddy-correlation method.
In this work, two mathematical tests to quantify the state of stationarity of time series obtained during eddy-correlation measurements, are presented and compared with each other using different data sets.

3.6 Corrections to flux measurements of reactive trace gases by enclosure techniques

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When using the enclosure techniques, observed fluxes of reactive trace gases must be corrected for those reactions which may occur in the air sample while passing through the enclosure (e.g. dynamic chambers) or remaining in the enclosure (e.g. static chambers). By proper design, selection of enclosure material and/or operational mode, losses of reactive trave gases to the inner walls of the enclosure can be minimized (or deliberately controlled). However, if enclosures are operated using ambient air, gas phase reactions of the involved compounds inside the enclosure can not be avoided. In case of the NO-NO2-O3 triad and under high ozone mixing ratios (> 50 ppb) and comparatively low NOX mixing ratios (< 5 ppb) such corrections can easily reach 50 % (and more) of the NO soil emission flux.
Using the mass balance equation of an enclosure, the individual corrections on the desired emission/deposition flux of a reactive trace gas are identified and corresponding formulations for their quantification are derived. Applicability of these formulations will be demonstrated on NO-NO2-O3 flux measurements by dynamic chambers in temperate and tropical environments. Finally, the developed concept of flux corrections will be extended to a "virtual" enclosure, the (inversion capped) in-canopy air space of a forest ecosystem. Under this conditions, a considerable part of the soil emitted NO may react with ozone (from aloft), forming NO2. Since NO2 uptake by plants (and soil) is much more effective than NO uptake, only a fraction of NO emitted from soils reaches the atmospheric boundary layer and the free troposphere. This effect, usually termed the “canopy reduction factor” (CRF), was repeatedly shown to reach 100 % (in terms of NO; up to 60 % in terms of NOX) in tropical primary rain forests, and -very recently-also in a mid-European forest ecosystem. Consequences for up-scaling those data of soil biogenic NO emissions , which have been measured on the forest floor, will be discussed.

3.7 (P14) Comparison of different coordinate systems for eddy covariance measurements

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A three-axis sonic anemometer-thermometer is the key instrument within a modern eddy covariance set-up for measuring atmospheric turbulence and the related fluxes. The wind vector measurement is obtained in the coordinate frame of the anemometer, and in most cases the observation must be first transformed into another frame, for both theoretical and practical reasons. Even in the case of level terrain, it is difficult to align the instrument with sufficient accuracy with respect to the local mean streamline, and a common practice is to apply a coordinate rotation to correct for any tilt. A traditional approach is to determine the so-called natural coordinate system from the wind data separately for each averaging period and perform either two or three rotations of the frame. Recently, it has been suggested that the so-called planar fit method, in which a fixed coordinate frame determined from an ensemble of observed mean wind vectors, would be a more appropriate choice for point measurements. In this paper we outline these methods and compare turbulent fluxes calculated in different coordinate frames. The eddy covariance data for the calculations are collected at three micrometeorological measurement sites of the Finnish Meteorological Institute. We discuss also the practical aspects related to the implementation of different methods.

3.8 (P15) Correction of surface flux measurements over heterogeneous terrain in case of horizontal advection

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Fluxes measurements over heterogeneous terrain such as urban areas, need to be performed at a quite high level above the surface in order to be representative of a large sample of individual elements composing the surface. Over such terrain, the blending height, which defines the lowest level of the inertial layer, can be higher than twice the canopy mean height. This requirement increases the problem of flux divergence between the surface and the measurements height. In case of meso-scale heterogeneity (coastal area, transition between two different ecosystems), horizontal gradients of the parameter of interest generate advection terms which lead to vertical divergence of the flux. The latter term can be quantified through the integration between the surface and measurement height of the budget equation of the parameter of interest (e.g. temperature for the sensible heat flux or specific humidity for the latent heat flux, etc.). The horizontal advection term has been computed for a case study during the field campaign CLU-ESCOMPTE over Marseilles (France). The city of Marseilles is situated on the Mediterranean coast and a flux measurement site was placed 2 km inland. Instruments were fixed at the top of a 30m tower installed on a terrace roof. Around this site, a network of temperature and humidity sensors had been set up and enabled the computation of horizontal gradients and thus advection. These terms have also been evaluated independently using high resolution (250m on horizontal) numerical simulation of a 3-day period with the Meso-NH atmospheric model using a specific parameterisation of the energy budget for urban areas. The results show that in this context of a coastal city, advection of heat is between 50 and 100 W m^{-2}. This is between 10 and 20% of the sensible heat flux. Impact of moisture advection is even higher on measured latent heat flux, since this term is very low over urban areas. The agreement between the estimates from measurements and from model output is very good for sensible heat flux divergence and more variable for the latent heat one. The methodology can be applied on any other parameter of interest.

3.9 (P16) Heat transfer during high wind episodes in the marine surface layer.

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The exact estimate of heat fluxes is of fundamental importance for applications of models at different scales. Therefore, a reliable subgrid parameterisation of these fluxes is needed. In the last few years, many scientists have addressed the limitation of the Monin Obhukov (M-O) theory in the surface layer, especially in the marine environment. Some of the factor that have been taken into considerations are non-local effects i.e. entrainemnet from large eddies Johanson (2003), waves, or presence of sea spray particles. This issue was recently taken into consideration by Andreas and De Cosmos (2002). They have reconsidered the question of the role of the sea spray in the transfer of latent and sensible heat by re-analysing data collected during the HEXOS (Humidity Exchange Over the Sea) experiment at the Dutch research platform Meetpost Noordwijk in the North Sea. Sea spray was found to contribute up to 40 % of the total latent flux for wind speed between 15 $ms^{-1}$and 18 ms^{-1}.
To estimate heat fluxes, eddy covariance method is applied using sonic anemometers for wind speed and temperature fluctuations. An unresolved problem is due to a systematic error that this kind of instrument suffer when measuring at high wind speed (reference..Grelle PhD thesis). Measurements obtained by sonics in Antarctica during high-speed katabatic flow show that negative heat fluxes can reach up to - 300 Wm^{-2} while profile method parameterisation shows fluxes not higher than -100 Wm^{-2}. There, one can observe a clear increase of heat flux values as wind speed increases and a negative flux of such a magnitude gives immediately raise to doubts on the correctness of the measurements. Although there can be few reasons to consider correct high negative heat fluxes during those episodes (the failure of the flux profile relationship due to blowing snow, which has the same effect as sea spray introducing sink and source of heat are; or the mixing of warmer air flowing above the ice during katabatic vents). One of the question to investigate is then how much of the high wT can be explained by instrumental error.
Here, we present examples from datasets in different environments i.e. Antarctica, marine and coastal stations.

3.10 (P17) Uncertainties in estimation of Soil CO_2 fluxes: Calibration of a Soil Respiration System

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Soil CO_2 efflux can be measured using several different chamber techniques, the most frequently used being closed static chambers, closed dynamic chambers and open dynamic chambers. These chambers affect the measurement process with each chamber type having its own limitations. Pressure anomalies, rising CO_2 concentration within the chamber and other factors affect the CO_2 gradient in the soil, which will ultimately affect the flux measured by the gas analysers. No single method has been established as a standard because methods have seldom been compared with known CO_2 effluxes, i.e. calibrated in an absolute manner. However, comparison against known CO_2 effluxes is the only way to standardize systematic errors of the different systems used for measuring soil respiration. Our experiment reports such a calibration procedure and the variability in measurements of soil respiration that can result even when using systems manufactured by the same company. The calibration system used in which a known CO_2 flux is generated through a layer of quartz sand. By varying the internal concentration of CO_2 beneath the sand, and also the particle diameter of the sand, a range of flux values typical of soil respiration can be reproduced. Chamber heads (referred as System A and B) to be tested are then inserted into the sand and the *apparent flux* recorded and plotted against the *actual flux.* System A produced values close to the actual flux if a soil collar was inserted into the sand. Without the collar there was a significant overestimation of the actual flux. In contrast, System B produced values close to the actual flux whether a soil collar was used or not. However, overestimation of the actual flux values still occurred when fine sand was used as the diffusion medium. It is obvious therefore that any overall calibration should incorporate a wide range of soil particle size and moisture content. Such calibration equations are established in for each of the soil respiration systems. The derived equation for System A accounts for 84% of the variance of the data, whilst that for System B accounts for 94%. The use of these two equations in transforming observed respiration data to a more real and accurate data in calculating the carbon loss through soil respiration in the Ecosystems.

4 Session: New Instruments

4.1 Application of aerosol mass spectroscopy for eddy-covariance measurements of speciated particle fluxes

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Measurements of surface / atmosphere exchange fluxes of individual aerosol components are needed (a) to quantify dry deposition inputs of NH_4^+, NO_3^- and SO_4^{2-} into sensitive ecosystems, (b) to improve our understanding of removal rates of aerosols from the atmosphere, as a function of surface roughness, particle size and composition, (c) to study gas-to-particle conversion and secondary aerosol formation, (d) to quantify emissions from source regions, such as urban environments. So far, only few micrometeorological flux measurements of individual aerosol components have been made, mainly using filter-packs in gradient configuration. This approach is laborious and non-continuous, prone to contaminations and relies on similarity theory. Here we present a novel eddy-covariance system, based on an Aerodyne Aerosol Mass Spectrometer (AMS), that is capable of measuring continuous fluxes of non-refractory aerosol components contained within PM_1, which includes NH_4NO_3, $(NH_4)_2SO_4$ and most organic carbon. The operating principles of the AMS are described, together with the modification, which have allowed this approach to be used for eddy-covariance. Example measurements are presented for two urban environments (Boulder, Colorado and Gothenburg, Sweden) and a forest (Duke Forest pine plantation, North Carolina). These data are used to demonstrate the capabilities and limitations of the current system and to motivate plans for future development: the current system is capable of resolving fluxes of 2 ng m^{-2} s^{-1} and deposition velocities of about 1 mm s^{-1} under the conditions found at the forest site. The measurements indicate that organic particulate matter and SO_4^{2-} were formed within and/or above the forest canopy, while NO_3^- was continuously being deposited. The urban measurements from Boulder identify the urban area as a source not just for hydrocarbon-like organic aerosol, but also for NO_3^- and oxygenated organics. It remains currently uncertain if these usually secondary compounds are emitted directly or formed through fast chemical conversion within the urban air space. By contrast, urban fluxes of SO_4^{2-} are negligible.

4.2 (P18) A first Step towards direct measurement of vertical fluxes of atmospheric particulate compounds

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Laser-desorption ionisation combined with time-of-flight mass spectrometry (LDI-TOF-MS) is a powerful tool to analyze individual atmospheric particles almost instantaneously. We present a methodology and a first experiment to combine disjunct eddy covariance measurements and single particle analysis in order to determine the flux direction of individual particulate compounds.
In May 2002, a field experiment was carried out to measure particle number fluxes and the chemical composition of ambient single aerosol particles at the forest ecosystem research site *Waldstein* in the *Fichtelgebirge* mountain range (50°09' N, 11°52' E). An eddy covariance system to measure vertical particle fluxes was mounted on a swinging boom 22 m above ground on a 30 m research tower. A YOUNG 81000 ultrasonic anemometer was employed. Next to the tower, the time-of-flight mass spectrometer (TOF-MS) LAMPAS 2 was operated for on-line analysis of the chemical composition of individual aerosol particles. The particles were directly introduced into the mass spectrometer through stainless steel tubing ending next to the measuring region of the ultrasonic anemometer. Particle sizing was accomplished through time-of-flight measurement of the aerodynamic particle diameter. This information was also used by an active-triggering circuit to ionize detected particles through laser desorption ionization (LDI). Data acquisition was synchronized with 10 Hz particle flux system using a DCF-77 radio-controlled clock.
Mass spectra of individual atmospheric particles can be separated depending on the vertical wind velocity at the time when the particle entered the mass spectrometer. This procedure yields virtual reservoirs of updraft and downdraft particles where each particle is correlated with a specific vertical wind vector. This approach is similar to disjunct eddy sampling and virtual accumulation. The virtual updraft and downdraft reservoirs can be analyzed separately and compared with each other. One problem with laser desorption ionization is quantitative analysis. There is no clear correlation between the concentration and the mass spectral peak area of a specific particulate compound. We reduced inter-particle variability by normalizing specific peak areas with the total peak area, and by averaging mass spectra of several particles. We applied a combined both concepts and extended the averaging procedure applying a Monte-Carlo-type simulation to obtain a statistical interpretation of mass spectra of a particle population. In this way relative concentrations of particulate compounds can be determined for different particle populations with a certain confidence. To determine the flux direction of a specific chemical compound such as NO_3^-, we compare the confidence intervals of the relative concentrations of this chemical compound in the

virtual updraft and downdraft reservoirs, respectively. Further development of this method has been proposed to the German Science Foundation (DFG).

4.3 (P19) Measurement of N_2O fluxes with eddy covariance using a quantum cascade laser absorption spectrometer

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A quantum cascade laser absorption spectrometer (Aerodyne Research Inc.) capable of measuring N_2O/CH_4 (at wavelengths of about 7870 nm) and NH_3 (at wavelength of 10310 nm) was combined with a sonic anemometer (METEK) to measure fluxes from fields. This equipment was used to measure N_2O and CH_4 fluxes from a maize field. The results show emissions of the order of 0.1 μg N_2O m^{-2} s^{-1}. The CH_4 flux was too low to be detected during this period.

4.4 (P20) Measurement of N_2O and CO_2 Based on Pulsed Quantum-Cascade Lasers

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Quantum-cascade lasers (QCLs) have passed from research to industrial production since their development 10 years ago. They are small, reliable, tunable and can be matched to a range of strong resonance lines of spectroscopically interesting molecules. QCLs driven in pulsed mode may be cooled thermoelectrically, which makes them attractive for high precision environmental applications.

Measurements were made with an instrument by Aerodyne Research Inc., which employs thermoelectrically cooled cascade lasers and detectors [1]. The QCL (Alpes Lasers, Switzerland) was driven around 4.46 µm with short (~ 10 ns) pulses and a 1% duty cycle at a temperature of approximately -25 °C. Extractive samples were measured at 65 mbar in a 0.5 l astigmatic multipass absorption cell with a path length of 56 m. The data acquisition system, TDL Wintel [2] controls the electronics and laser driver, monitors the IR detector, and analyzes the spectra to derive mixing ratios based on spectral parameters from HITRAN [3]. Spectral scans are obtained by a sub-threshold voltage ramp which creates a bias temperature. The relevant parameters were chosen to allow simultaneous detection of N_2O and CO_2 at ambient concentrations.

Laboratory tests showed excellent linearity for N_2O and CO_2 in the concentration ranges 0-1000 ppb and 0-1000 ppm, respectively. There was no experimentally detectable cross-sensitivity to CO_2 or H_2O, which is especially valuable for N_2O measurements that are often limited by overlapping absorption spectra when using NDIR techniques. Quantification based on HITRAN parameters differed by up to 20 % when compared to certified calibration gases. This can be overcome by regularly measuring a reference gas. After stabilization of the optical bench and the pulse electronics we obtained a minimum Allan variance corresponding to 0.12 ppb (0.05 %) or 0.56 ppm (0.19%) for N_2O and CO_2, respectively.

The instrument can be used without cryogenic cooling for long-term, continuous measurements with little maintenance. High precision ambient air monitoring and validation has been performed employing an optimized sequence including background and reference gas measurements. In combination with a strong vacuum pump, a time resolution of 10 Hz can be obtained which makes it suitable for Eddy correlation flux measurements.

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[2] Nelson, D.D., Shorter, J.H. et al. (2002), *Applied Physics B,* 75, 343-350.
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4.5 (P21) A Fast Response Chemiluminometric Ozone Sensor for Atmospheric Applications.

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The standard method of ozone in the atmosphere is the absorption of UV light at 254nm and this is the mainstay of most commercial instrumentation. UV photometry does however require a relatively long integration time to attain a good limit of detection (LOD, S/N = 3) of 1 ppbV, this results from a data rate in the region of 10 seconds. Whilst this is perfectly acceptable for air quality monitoring where hourly averages may be the goal of the measurements it is not ideal for research on small scale atmospheric phenomena. Some chemical reactions produce light as excited state intermediates relax to the ground state, this phenomena is known as chemiluminescence, this is the mainstay of atmospheric measurement of NOx. Chemiluminescence (CL) is an applicable method for the determination of ambient ozone particularly where there is a requirement for faster data rates, such as aircraft studies and flux measurements. Flux studies are invaluable for reconciling sinks and sources of atmospheric tracers. Ozone deposition velocities in particular contribute to some significant differences between atmospheric models.

Recently a study has described the use of photoactivated chromotropic acid for the detection of ozone in the atmosphere. Preliminary field trials showed that the system had a detection limit of less than 1 ppbV, a sensitivity of 100 pptV based on a sample rate of 2 Hz. This clearly shows great promise as a novel method for the fast determination of ozone in the atmosphere, in particular for flux studies. Modern PMTs mean that a small sensor package can be produced, it is expected that the overall instrument package be less that 2 kg including data system, with very low power consumption. The size of the package makes it a potential addition to tethersondes and also to UAV platforms. These platforms provide a very low cost alternative to research aircraft which not only very expensive are limited in the areas they can operate. Small UAVs capable of carrying such a sensor do exist and have already been used for chemical measurements ion the atmosphere. They are not subject to the increasing air traffic strict rules.

This instrument will be deployed alongside a commercial fast response open path H_2O/CO_2 sensor and a standard sonic anemometer which will provide an excellent comparison with scalar fluxes and form the basis of suite of trace gas flux measurements. It is envisaged that intended deployments include a forthcoming SOLAS shipcruise and a forest canopy flow experiment.

4.6 (P22) Ammonia Eddy Flux Measurements by QC-TDLAS during TORCH-1

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Around 90% of ammonia emissions in the UK are produced by agricultural activities (particularly livestock and fertilisers), but urban and related sources are becoming significant. Reduced nitrogen is the main contributor to the total nitrogen deposition in the UK and so understanding its emission and deposition to terrestrial systems is important. This relies on accurate measurements of ammonia fluxes from both natural and urban sources. A novel Quantum-Cascade Tuneable Diode Laser Absorption Spectrometer (QC-TDLAS; Aerodyne Research, Inc.) was deployed as part of the first Tropospheric Organic Chemistry (TORCH-1) Experiment to measure ambient ammonia and NO_2 concentrations. In addition, eddy flux measurements were attempted with this system. TORCH-1 was a large UK field experiment designed to increase understanding of the role of primary and partially oxidised organic species in atmospheric gas phase photochemistry. This is being achieved by development of thermodynamic and microphysical models of organic aerosol behaviour, which are being validated against field data from "near" and "far-field" plume data downwind of London. TORCH-1 was conducted during July and August 2003, at Writtle Agricultural College in Essex, a semi-rural location NE and immediately downwind of the London pollution plume. Ambient concentration measurements were provided by the Peltier cooled QC-TDLAS system operating at 967 cm^{-1} and 1606 cm^{-1} respectively for NH_3 and NO_2. The system was installed overlooking a large wheat field with canopy height between 80-100 cm. The field was harvested during the experiment after which the main body of ammonia flux measurements were subsequently obtained. Eddy flux measurement results will be presented and discussed and the performance of the system is examined in relation to lab based inlet response tests and how the wall interactions impact on the flux measurements. Finally the use of the TDLAS measurements to study equilibrium of ammonia gas-to-particle conversion states during TORCH-1 to assess the aerosol composition state in the London plume are also discussed in combination with high resolution measurements of aerosol composition by an Aerosol Mass Spectrometer (AMS).

4.7 (P23) CLASP - a Compact Light-weight Aerosol Spectrometer Probe.

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The need for an accurate Source Function of the production of Sea Spray Particles under a range of environmental conditions has been emphasised by recent global climate studies. Sea spray particles are the second largest single source of aerosol mass injected into the atmosphere. They are particularly important as a source of cloud condensation nuclei (CCN), thus they affect the chemistry and radiative effects of marine clouds. This is of specific interest as marine stratocumuli are a major unknown in climate studies. Sea Spray Particles also play a large role in transferring heat and moisture across the air-sea interface and limit the range of military equipment by scattering the infrared radiation used in their optical surveillance equipment. Work over the last 30 years has reduced the uncertainties in the function from 7 orders of magnitude to just 1 but greater precision is required for the new generation climate models. This has been made possible by recent advancements in instrumentation and techniques.
The poster will explain all about the new instrumentation CLASP (Compact Lightweight Aerosol Spectrometer Probe) that's being developed at the University of Leeds and will describe why it is particular useful as a particle counter for flux measurements of sea spray. With international collaborators, we are trying to derive Sea Spray Source Functions applicable to the world's oceans by using a variety of locations. Some of results from a couple of field test at different locations will be shown.

4.8 (P24) Model experiments and analyses of burning products of different heating materials and fuels

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Chemical processes and cycles in the atmosphere as well as disastrous pollutants, which generate evident and still obscure climate changes, are serious tasks for scientists around the whole world. Scientists from the Department of Analytical Chemistry for almost seven years are implicated in air pollution problems. There are constructed several simple and more complicated systems for aerosol sampling from close box, in which different types of fuels were burned in the laboratory conditions. In the current work various methods of sampling, sample preparation and analyses were developed to expand our knowledge in such a way that the fate of pollutants can be fully understood.

Today the understanding of burning processes in which the main part of pollutants is produced is a very consequential question. That is why in the Department of Analytical Chemistry several convenient systems have been made for the burning of different fuels and heating materials. The simple system, which consists of the close box, the sampler, air pump and flow meter, was constructed for burning paraffin and stearin candles. These experiments showed that it is possible to find convenient burning conditions to obtain aerosol particles and black carbon of different concentration levels. In the next step burning systems were adapted to burn fuel, diesel fuel and kerosene. We tried to burn kerosene in the previous system, but obtained particles were very large in size and it let conclude that the previous system was not suitable for burning of kerosene. In obtained aerosol particles using Smokestain Reflectometer M 43-D black carbon was detected. Some heavy metals were determined by stripping potentiometry and atomic absorption spectrometry and X-ray spectrometry was used to analyse Al, P, Mn, Ni, Fe, As and etc. In the current experiments kerosene with known, different spikes of tetraethyl lead was burned and in the obtained aerosol particles correspondent amount of lead was determined. It may to conclude that used fuel burning and sampling methods as well as the sample preparing method was applicable.

The next step of our scientific work was experiments with car exhaust, which was capture directly from the car. A convenient system that was equipped with air pump and flow meter was constructed for car exhaust capturing. In current experiments cars with different kind of engines and of various brands were chosen. In the sampled aerosol particles from car exhausts black carbon and heavy metals were determined. Attained results shows that lead are even 10 times higher in diesel fuel than in fuel, black carbon concentration also is much higher in diesel fuel and these particles are bigger in size than particles from fuel.

4.9 (P25) High Precision Trace Gas Analysis by FTIR Spectroscopy

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The three most important anthropogenic greenhouse gases are carbon dioxide, methane and nitrous oxide. While not directly a greenhouse gas, carbon monoxide also plays an important role in atmospheric chemistry as one of the most important factors determining the concentration of the atmospheric oxidants [1]. There are, however, large uncertainties in their sources, and due to the importance of these gases better estimates of their emissions from various ecosystems are required.
We developed a method based on a portable Fourier Transform Infrared (FTIR) spectrometer which allows the simultaneous on-site analysis of multiple trace gases (CO_2, CH_4, CO, N_2O) as well as the $^{13}C/^{12}C$ isotopic ratio of atmospheric CO_2 (i.e. $\delta^{13}CO_2$). The method requires no sample preparation other than optional drying and can be applied directly to ambient air samples. Precision strongly depends on the desired time-resolution. While a maximum temporal resolution of 1Hz is possible [2], maximum precision is obtained with an averaging time of 5 min. Applicability of our approach, for ambient air monitoring was demonstrated during an inter comparison study with independent measurement techniques like GC-FID/ECD and a pulsed quantum cascade laser (QCL). Extractive FTIR spectroscopy has the necessary precision, time resolution and transportability to be used for micrometeorological flux-gradient techniques. Furthermore, the ability to continuously determine $\delta^{13}CO_2$ bears the potential to study processes with distinctive isotopic signatures.

[1] A.M. Thompson, Science, 256 (1992) 1157-1165.
[2] J. Mohn, A.-M. Forss, S. Brühlmann, K. Zeyer, R. Lüscher, L. Emmenegger, M. Weilenmann and N.V. Heeb, International Journal of Environment and Pollution, (2004)

4.10 (P26) Comparison of fluxes measured with METEK USA-1 sonic anemometers using 2D and 3D sensor head correction

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A sonic anemometer is normally the core instrument of all flux measurements and flow distortion around the sensor head is a know problem that must be handled for all sonic anemometers. Many sonics have built in algorithms for an on-line 2 dimensional (2D) sensor head correction of the measurements and now the METEK USA-1 “Scientific” sonic anemometer also provides a 3 dimensional (3D) sensor head correction.
The 3D correction is expected to be important under strong convective conditions, in complex terrain and above rough canopies like forests. In all these situations the vertical and the horizontal components of the flow can be of the same order and the use of a 3D sensor head correction seems to be very sensible.
The new 3D correction was developed when the new processor generation was introduced to the METEK USA-1 called “Scientific”. The algorithms are found from wind tunnel measurements and allow corrections over the complete 0 … 360 ° range in azimuth and within +- 45 ° zenith angle. For the “Scientific” version the 3D correction can be applied on-line during the measurements. But for all Metek sonics with the same sensor head design the 3D correction can also be applied off-line on already recorded data.
We will show examples of and compare the result from eddy correlation flux measurements of momentum and sensible heat calculated with both the 2D and 3D sensor head corrections.

5 Session: Extension from point to area

5.1 Review of the current knowledge on footprints.

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The area contributing a certain fraction to the measured flux is called the footprint or the source area of the measurement. For a given geometry (observation height, roughness length, canopy structure) and meteorological situation (wind speed and direction, intensity of turbulence, stratification) the footprint function, also called a source weight function connects the actual sources on the surface to the measurement at the observation point. The determination of the footprint (function) is not a straightforward task and several theoretical approaches have been developed. They can be classified to four categories:

- Analytical models
- Lagrangian stochastic trajectory models
- Large eddy simulations
- Ensemble-averaged closure models

The models need experimental verification and probably the most used method is based on the release of the trace gases, but again, the planning of the perfect experimental set-up is not a straightforward task.

The main future directions could be:

- to improve understanding on canopy turbulence and stable conditions
- to develop easy-to-use parameterizations for footprint estimates
- to improve the description of horizontal inhomogeneities and non-flat topography
- to develop models for active scalars (chemically reactive gases, aerosol particles)

Rferences:

Baldocchi, D.: 1997, Flux footprints within and over forest canopies, Boundary-Layer Meteorol., 85, 273-292.
Schmid, H.P., 2002. Footprint modeling for vegetation atmosphere exchange studies: a review and perspective. Boundary-Layer Meteorol., 113, 159-183.
All articles in Special Issue: Flux and Concentration Footprints. Agricultural and Forest Meteorology 127, 2004.

5.2 Footprint for dummies

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Flux measurements with micrometeorological methods are determining the fluxes where the sensors are physically located. Often the main interest is not this flux, but the exchange flux of an ecosystem under investigation for which the measuring system has been installed. From a pragmatic point of view a tool is needed that evaluate to which extent a specific area is contributing to the measured flux.
The footprint of a flux measurement can be understood as a probability density function that describes how strongly a surface exchange at any location upwind of the sensor affects the measured flux. In order to calculate footprints for flux measurements the analytical footprint model developed by Kormann and Meixner (2001) was applied. It is able to describe two dimensional footprints for receptors within the surface layer under non-neutral conditions and is simple enough to be routinely applied for the analysis of flux measurements. We developed an EXCEL version that allows calculating footprint contributions for different fields based on Sonic anemometer data.

Kormann, R., and Meixner, F. X.: An analytical footprint model for non-neutral stratification, Bound.-Layer Meteor., 99, 207-224, 2001.

5.3 Aggregation modelling

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Aggregation is here defined as non-linear area-averaging of surface fluxes from local scale to regional scale. Important non-linear effects are included in the scheme of the micro-scale aggregation model. It is a physically based approach balancing the advective term and the diffusive term of the atmospheric flow equation. Atmospheric statitic stability is parametrized with Monin-Obukbov similarity. The model equations are linearized and solved by Fast Fourier Transform, a very efficient computational solution. The atmospheric flow is continuously adjusting itself as a function of the characteristics of the surface such as roughness, temperature, vegetation and humidity variations in the landscape. A series of internal boundary layers develop in heterogeneous terrain and the added effect of these is quantified through the model.
Input to the model are 1) wind vector and air temperature at a height above the surface where the significance of each individual internal boundary layer is minor, i.e. at the blending height, and 2) high resolution satellite maps of the lower boundary conditions. These typically comprise a land cover map, a surface temperature map, a vegetation density map (leaf area index estimated through e.g. the Normalized Difference Vegetation Index) and a roughness map. The roughness map may either be a simple assignment of local roughness per land cover type, or a combination of satellite-based crop height maps and relationships between canopy height and roughness for specific crop types.
Surface momentum and sensible heat fluxes are calculated directly from the micro-scale aggregation model per grid cell, e.g. of the order of 10 to 30 m by 10 to 30 m. The high-resolution results contain the local-scale information including the non-linear turbulent responses between each individual patch within the terrain as a function of fetch, position and height. The atmospheric flow model ‘remembers’ the flow through the landscape in true terrain coordinates, i.e. no grouping of tiles is performed. Hence the model results can used to compare field-scale observations to larger scale values through the high-resolution surface flux results.
The surface flux results typically are aggregated for areas of ~1 to 15 km by 1 to 15 km (regional scale). The aggregated values of the surface fluxes allow calculation of the effective roughness for momentum and heat ($<z_0>$ and $<z_{0t}>$) for the specific type of landscape and atmospheric condition. The effective roughness values are those that represent the average conditions of the heterogeneous landscape. The effective roughness is useful in larger-scale grid models (meso-scale models, weather forecasting, climate and agro-hydrological models).
The basic equations, the model structure as well as results from various heterogeneous landscapes are presented.

Further reading:

Hasager, C.B. Nielsen, N.W., Jensen, N.O., Boegh, E., Christensen, J.H, Dellwik, E. and Soegaard, H., 2002 Effective roughness calculated from satellite-derived land cover maps and hedge information used in a weather forecasting model. *Boundary-Layer Meteorology* 109, 227-254,

Hasager C.B. and N.O.Jensen, 1999, Surface flux aggregation in heterogeneous terrain, *Quart. J. Royal. Meteorol. Soc.* **125**, 2075-2102

5.4 From the leaf to the atmosphere: Mechanistic studies to investigate biosphere-atmosphere exchange

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Substantial amounts of trace gases such as volatile organic compounds, nitrogen oxides and reduced sulphur compounds are exchanged between biosphere and the atmosphere. Within these processes chemistry, physics and biology are closely coupled. For interpretation, upscaling and diagnosis, modelling is necessary. It should be supported by understanding production/consumption and exchange regulations on a leaf or branch level. Some data will be presented to demonstrate this need. During recent field experiments we found European beech to act as a high monoterpene emitter related to a light/temperature regime. Introducing this information into modelling studies, a significant impact on emission budget calculations could be demonstrated. Furthermore, understanding the close relation between primary productivity and trace gas emission helped to evaluate seasonality and plant development demonstrated by results obtained with Amazonian tree species. Investigations of the close relations between the plant uptake of CO_2 and carbonyl sulphide (COS) resulted in a new global estimate of the COS sink strength. Better knowledge on compensation points helped to assess the bi-directional exchange of trace gases, such as NO_2 and COS. These few examples may demonstrate the relevance of mechanistic studies.

5.5 Regional CO_2 and H_2O budgets and fluxes over homogeneous and heterogeneous land surfaces

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The uptake of CO_2 by vegetation was studied experimentally in June 2001 on a regional scale of several hundreds of square kilometers using a Lagrangian budgeting approach. For this purpose, CO_2 and meteorological parameters were measured from a small aircraft over a flat homogeneous and productive temperate forest in the Landes region, south of Bordeaux. During one flight with perfect Lagrangian conditions an average midday (12:30 - 14:30 UTC) forest CO_2 uptake rate of 16 ± 2.5 µmol m^{-2} s^{-1} was observed. The derived CO_2 flux for the experimental area was about 15 % smaller than the local net ecosystem exchange (NEE) measured by eddy covariance at a tower north of the flight domain, and about 12 % higher than model estimations, which were based on remote sensing (satellite) data. The contribution of anthropogenic emissions to the regional CO_2 budget was estimated from simultaneous CO measurements on the same platform to be less than 0.5 µmol m^{-2} s^{-1} (Schmitgen et al., 2004).
Secondly, the impact of local differences in surface structure and land use on the spatial and temporal distribution of greenhouse gases in the near surface boundary layer was investigated during an airborne observation campaign in August 2004. Atmospheric concentrations of gases (H_2O, CO_2, CO, NO, NO_2, NO_y) and several meteorological parameters were measured with high spatial and temporal resolution over intensively managed farmland of the Jülicher Börde. Additionally a hyperspectral scanning system was deployed downlooking to measure spatio-temporal variations of photosynthesis at canopy level, in order to quantify the biological activity of the vegetation. Preliminary results suggest that surface structures were mapped into the distribution of CO_2 and H_2O, showing pronounced small scale patterns in both, concentrations and fluxes, and in the correlation between the measured anthropogenic and biogenic trace gases. The correlations can be used to identify and separate biological (photosynthesis and transpiration) and anthropogenic activities (traffic, industry and power plants), allowing to quantify the contributions of different sources and sinks to the CO_2 and H_2O budgets.

Schmitgen, S., Geiß, H., Ciais, P., Neininger, B., Brunet, Y., Reichstein, M., Kley, D., Volz-Thomas, A. (2004): Carbon dioxide uptake of a forested region in southwest France derived from airborne CO2 and CO measurements in a quasi-Lagrangian experiment. - J. Geophys. Res. 109, D14302: doi:10.1029/2003JD0043