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TURBULENT SURFACE FLUXES AND ECOSYSTEM  
FUNCTIONING IN BOREAL CONIFEROUS FORESTS

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Academic dissertation

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## Abstract

Interactions between the atmosphere and the Earth's surface are key issues in global climate change. Terrestrial ecosystems are at the centre of these processes because they act as sources and sinks of important greenhouse gases and of climate-cooling aerosol particles. The aim of the present work was to improve our understanding of the interactions between the atmosphere and boreal forests. For this purpose, we analysed turbulent surface fluxes of trace gases and aerosol particles measured with eddy-covariance (EC) technique during 1996—2002 and studied their connection with ecosystem functioning. In addition, we utilised supporting measurements of environmental conditions. Most of the data used in this study were obtained from the SMEAR II field station in Hyytiälä, southern Finland.

EC is a direct micrometeorological flux measurement method, based on the covariance between vertical wind speed and a variable of interest, such as gas concentration or temperature. It is a useful tool for studying the spatial and temporal variability in surface fluxes which, in turn, reflect the functioning of a whole ecosystem in the scale of about 0.1 to 1 km. However, typical error sources such as night-time periods of low turbulence must first be taken into account. Our analysis of EC fluxes revealed that the Scots pine forest in Hyytiälä was a sink of carbon, assimilating about  $200 \text{ g(C)m}^{-2}\text{yr}^{-1}$  on average with a variation of about  $80 \text{ g(C)m}^{-2}\text{yr}^{-1}$  or less. We also observed that both the daytime and the night-time  $\text{CO}_2$  fluxes contributed markedly to the variability in carbon exchange, indicating that photosynthesis and respiration have an equally important influence on the net ecosystem exchange of  $\text{CO}_2$  (NEE). The recovery of photosynthesis in spring had the best connection with air temperature, and our results suggest that higher spring temperatures will most likely lead to earlier spring recovery of photosynthesis and to longer growing seasons in some parts of the boreal zone. The effect of rising air temperatures on soil warming and, consequently, soil respiration appears to be more complicated and less direct than the effect on growing-season length.

Climatic factors had a clear seasonal influence on most surface fluxes. This was because cloudiness and time of year affected the amount of incoming energy and because radiation, temperature, and precipitation influenced the metabolic activity of the ecosystem. On diurnal scale, these environmental drivers had a direct effect on fluxes of  $\text{CO}_2$  and water vapour ( $F_C$  and  $F_w$ , respectively) and on the deposition velocity of  $\text{O}_3$  ( $v_{d,\text{O}_3}$ ) but no observable effect on the deposition velocity of fine particles ( $v_{d,p}$ ). On the other hand,  $v_{d,p}$  was directly affected by the physical amount of foliage whereas  $F_C$ ,  $F_w$ , and  $v_{d,\text{O}_3}$  were largely insensitive to it. This was evident after a thinning in which 25% of the tree stems were removed; our results suggest that the redistribution of trace-gas sources and sinks within the ecosystem was comprehensively able to compensate for the lower foliage area. Furthermore, we showed that although  $v_{d,p}$  appeared to have no clear connection with any climatic variable, it was higher during new-particle formation events, most likely because of the relatively larger amount of small, nucleation-mode particles. Finally, terpenoids, side products of photosynthesis, emerged as a link among forest metabolism, aerosol dynamics, and climate. Because of the role of forests as sinks of  $\text{CO}_2$  and as sources of aerosol particles, increased vegetation growth potentially leads to the slowing down of global warming.

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## List of publications

This thesis consists of an introductory review followed by five research articles. The papers are reproduced with the permission of the journals concerned.

I: **Suni, T.**, Berninger, F., Markkanen, T., Keronen, P., Rannik, Ü., and Vesala, T. (2003) Interannual variability and timing of growing-season CO<sub>2</sub> exchange in a boreal forest. *Journal of Geophysical Research*, 108: D9, 4265, doi: 10.1029/2002JD002381

II: **Suni, T.**, Berninger, F., Vesala, T., Markkanen, T., Hari, P., Mäkelä, A., Ilvesniemi, H., Hänninen, H., Nikinmaa, E., Huttula, T., Laurila, T., Aurela, M., Grelle, A., Lindroth, A., Arneth, A., Shibistova, O., and Lloyd, J. (2003) Air temperature triggers the recovery of evergreen boreal forest photosynthesis in spring. *Global Change Biology*, 9, 1410-1426

III: **Suni, T.**, Rinne, J., Reissell, A., Altimir, N., Keronen, P., Rannik, Ü., Dal Maso, M., Kulmala, M., and Vesala T. (2003) Long-term measurements of surface fluxes above a Scots pine forest in Hyytiälä, southern Finland, 1996–2001. *Boreal Environment Research* 8:4, 287-302

IV: Vesala, T., **Suni, T.**, Rannik, Ü., Keronen, P., Markkanen, T., Sevanto, S., Grönholm, T., Kulmala, M., Ojansuu, R., Uotila, A., Levula, J., Mäkelä, A., Pumpanen, J., Kolari, P., Berninger, F., Ilvesniemi, H., Nikinmaa, E., and Hari, P. (2004) The effect of thinning on surface fluxes in a boreal forest. *Global Biogeochemical Cycles* (resubmitted)

V: Kulmala, M., **Suni, T.**, Lehtinen, K.E.J., Dal Maso, M., Boy, M., Reissell, A., Rannik, Ü., Aalto, P., Keronen, P., Hakola, H., Bäck, J., Hoffmann, T., Vesala, T. and Hari, P. (2004) A new feedback mechanism linking forests, aerosols and climate. *Atmospheric Chemistry and Physics* 4, 557–562

## Author's contribution

I am fully responsible for the summary part of this thesis. In **Paper I**, I was responsible for all the analyses and for the writing of the article; in **Paper II**, for all the analyses and for a major part of the writing; in **Paper III**, for the writing and for all the analyses except for the determination of new-particle formation events; in **Paper IV**, for all the eddy-covariance data analyses, for a small part of the writing, and for editing; in **Paper V**, for a part of the writing and editing and for all the analyses except for the determination of  $J_3$  and GR.

# 1. Introduction

Current climate models predict an increase of 2 to 6 °C in global temperature during the next 100 years (*Climate Change* 2001). This rate of warming would be unprecedented in the Earth's history and would most likely have far-reaching ecological, economic, and societal consequences. In the models, however, biogeochemical feedbacks among the components of the climate system - land, ocean, and atmosphere - are a major source of uncertainty. Important interactive processes underlying surface-atmosphere exchange are still poorly understood, and the predictions remain highly uncertain. Carbon dioxide (CO<sub>2</sub>) and water vapour are greenhouse gases that warm the climate. Aerosol particles cool the climate both directly by reflecting incoming solar radiation back to space and indirectly by forming cloud droplets. A variety of biogeochemical interactions affect the concentrations of CO<sub>2</sub>, water vapour, and particles. Vegetation plays an important part by emitting volatile organic compounds (VOC's) such as terpenoids that may form aerosols through rapid reactions with ozone (O<sub>3</sub>) and other atmospheric oxidants (Calogirou *et al.* 1999, Atkinson 2000, Raes *et al.* 2000, Jenkin and Clemitshaw 2000). The products of these reactions have low volatility, and they readily take part in gas-to-particle conversion processes such as condensation. Global warming and CO<sub>2</sub> fertilization are likely to lead to increased photosynthesis and forest growth. This will increase forest biomass and emissions of VOC's and thereby probably enhance organic aerosol production (**Paper V**). Such feedback mechanisms and other interactions were recognised in the report by the Intergovernmental Panel on Climate Change (IPCC) in 2001 (*Climate Change* 2001). The report emphasised the complexity of the combined direct and indirect forcing from both aerosols and gases. Knowledge of the interactions between gases and particles would reduce the uncertainty in current estimates of radiative forcing and improve predictions of the effects of anthropogenic activity on global change. Terrestrial ecosystems as sources and sinks of particles and gases are in the centre of these interactions.

In recent years, eddy covariance (EC) technique has emerged as a direct way to determine the whole-ecosystem exchange for some gaseous species and for aerosol particles down to 10 – 20 nm in diameter. EC-based estimates of annual budgets of CO<sub>2</sub> exchange, for example, require measurements made throughout the year in all kinds of atmospheric conditions. However, because of systematic problems related to periods of low turbulence (typically during summer nights), these budget estimates can be susceptible to large errors. On the other hand, EC measurements made during well developed turbulence can accurately reveal ecosystem responses to environmental factors. Therefore, this technique is particularly suited for determining the response of exchange rates of whole ecosystems to environmental perturbations, the relationship between fluxes and phenology, and the factors causing year-to-year variability in fluxes. The magnitude and seasonal patterns of carbon and water cycles and particle production vary considerably with latitude and ecosystem type, and the significance of boreal ecosystems to global carbon cycling and particle production is still uncertain. In boreal conditions, the annual variability of most surface fluxes is extreme, and little information of annual behaviour and budgets can be gained from short-term measurements. Long-term field measurements of gas and particle fluxes encompassing periods of extreme conditions such as drought or very cold winters are therefore

essential, also for purposes of global change modelling (Grelle *et al.* 1999). Moreover, they are important for investigating the combined effect of respiration and photosynthesis on an ecosystem carbon balance at present and in the future.

## 2. Aims

Forest-atmosphere interactions are among the key issues in global climate change. The two most important drivers for change in the net ecosystem exchange of CO<sub>2</sub> (NEE) are the climate and natural or human-induced disturbances. Whether global climate warming will serve to increase or decrease the carbon sink in boreal forests is uncertain, since the warming may lead to longer growing seasons but also to increased soil respiration especially in the winter. The role of disturbances such as fires, land use change, and commercial management of forests may be more significant in forest-atmosphere interactions than has been assumed until recently, since the majority of EC measurements are performed in undisturbed ecosystems, giving a potentially distorted view of natural processes.

The aim of the present work was to improve our understanding of the interactions between boreal forests and the atmosphere by utilizing the EC technique in a versatile way in connection with supporting environmental measurements. Particular objectives were:

- To assess the seasonal and interannual variability in NEE in a boreal forest in Hyytiälä, southern Finland, and to determine the relationship between NEE and environmental drivers on diurnal and seasonal scale (**Papers I & III**)
- To take a step towards determining the effect of climate warming on the boreal carbon cycle by assessing the driving environmental factors of springtime photosynthetic recovery in boreal coniferous forests (**Paper II**)
- To assess the influence of climate and human-induced disturbance on fluxes of inert and reactive trace gases and particles in Hyytiälä (**Papers III & IV**)
- To determine potential interactions among climate, aerosol dynamics, and forests by assessing connections among biogenic terpenoids, oxidants, and particle formation and growth rates observed in the forest in Hyytiälä (**Paper V**).



### 3. Surface fluxes

#### 3.1. Atmospheric boundary layer

In the troposphere, the atmospheric boundary layer (ABL) is the part in which the Earth's surface directly influences the movement and properties of air. For instance, rough surfaces may cause mechanical turbulence in the overflowing air, and daytime heating of the ground may result in warm thermals rising within the ABL. Above the ABL lies the *free troposphere*, sometimes called the *free atmosphere* (Fig. 1), in which the effect of the Earth's surface is not as evident. The diurnal variation in the thickness of the ABL is large: At night, the ABL can be only a couple of hundred metres deep (*stable nocturnal boundary layer*), and by day it may grow up to three kilometres (*mixed layer*) (Stull 1988). The volume of the ABL affects all surface fluxes. In the deep daytime boundary layer, the concentrations of gases and particles are mixed effectively. At night, the boundary layer is shallow and mixing is less effective (Fig. 1). Consequently, the net emission or deposition of gases and particles has a larger influence on concentrations within and just above the forest at night than in daytime, potentially resulting in night-time accumulation or depletion of different gas and particle species. Concentration profiles may then change abruptly in the morning when the mixing begins again. This dilution of accumulated species or the replenishment of depleted ones in the morning, *the morning flush*, is especially important in boreal coniferous forests where the ABL often grows notably deep compared to temperate forests (see 5.2).

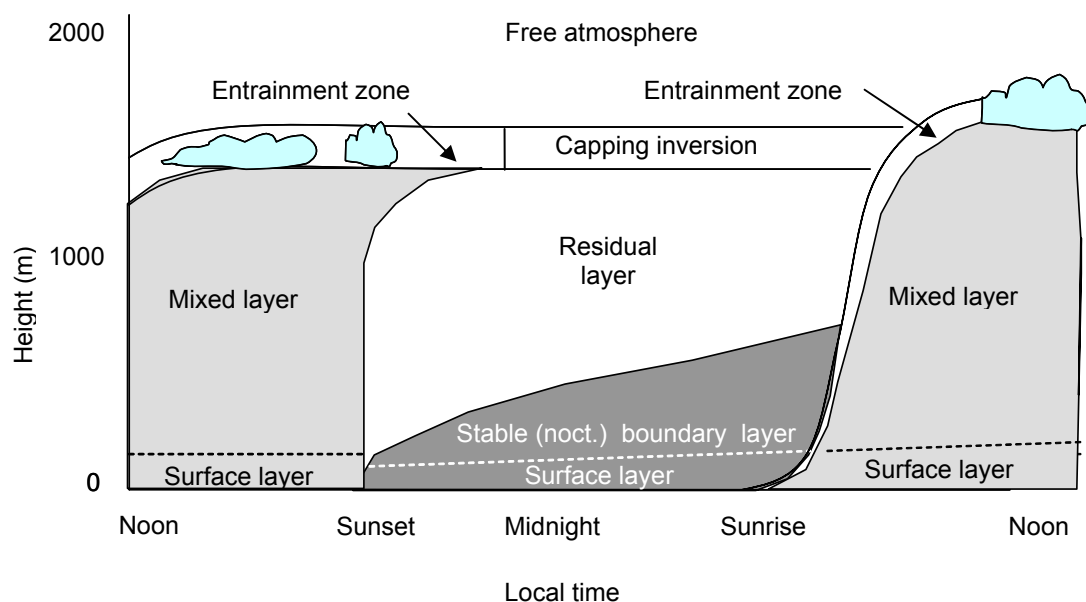


Figure 1. Diurnal development of the atmospheric boundary layer (ABL). During daytime the ABL consists of a very turbulent mixed layer (light grey). At sunset, a stable nocturnal ABL (dark grey) starts to grow from the surface upwards, and former mixed-layer air becomes a less turbulent residual layer (white). At sunrise, a new mixed layer starts to form at the surface, mixing new air into it from above at the entrainment zone. The surface layer is, by definition, the lowest 10% of the ABL. Adapted from Stull (1988).

### 3.2. Turbulent transport and surface fluxes

Atmospheric turbulence transports matter and energy to and from the Earth's surface within the ABL. We observe turbulence as the irregular gustiness of wind, as swirls that flow leaves up and down on sidewalks. These swirls are called *eddies* (Fig. 2). The size of eddies varies from less than a millimetre to several hundreds of metres. The intensity of turbulence can be described by *the friction velocity*  $u^*$ , given by

$$u^{*2} = \overline{|u'w'|} \quad (1)$$

where  $u'$  and  $w'$  are the fluctuations of horizontal and vertical wind velocity, respectively, with  $u'$  pointing in the direction of the mean wind. A practical way to interpret  $u^*$  is to think of it as the tangential velocity of circular, rotating eddies (Monteith and Unsworth 1990).

Turbulent transport in air is many orders of magnitude more efficient than molecular diffusivity and by far the most important transport mechanism between the atmosphere and the surface (Stull 1988, Rannik 1998). In dry conditions, gases and aerosol particles are removed from the atmosphere onto surfaces by *dry deposition* (Sehmel 1980). For gases as well as for particles, meteorological variables (friction velocity and roughness length), concentration gradients, chemistry, surface characteristics (stickiness, wetness, canopy structure), and different dependencies on climatic variables affect the deposition rates. For gas deposition, another important factor is stomatal control that reflects the physiological stage of vegetation. Of the properties of gases, the most important ones are chemical reactivity and water solubility that enter into the final removal process on the surface, especially if the surface is wet: In general, deposition on a water surface is limited by the solubility of the gas, but if irreversible chemical reactions consume the deposited gas molecules inside, for instance, a water film on a leaf, the concentration of the depositing species in the water does not increase and deposition can go on as long as the reactions take place (Sehmel 1980). For aerosols, the dry deposition depends largely on the physical and chemical properties of the particles (diameter, solubility) (Sehmel 1980).

The contribution of turbulent transport through the surface layer is similar for gases and particles, but for large particles, also sedimentation may enter into the process (Seinfeld and Pandis 1998). In the quasi-laminar sublayer, the transport occurs by diffusion and sedimentation. Finally, the uptake on the surface can occur by reversible or irreversible absorption (gases) or adhesion (particles). Dry deposition rates for gases depend also on the physiological stage of the vegetation: During night or winter dormancy (evergreen conifers), the stomata are closed and mass transfer is reduced, if not completely inhibited (Sehmel 1980). For deciduous trees, the effect is very pronounced because of the annual rhythm of leaf-out and leaf senescence. In addition, the deposition rate onto snow can be clearly smaller than on bare soil or vegetation (Sehmel 1980, Zeller 2000). The penetration of the pollutant through the canopy, be it gas or particles, is also affected by the structure of the canopy (Sehmel 1980).

The lowest part of the ABL where  $u^*$  and vertical turbulent transport are approximately constant with height (varying by less than 10%; this is true in the lowest 10% of the ABL) is called the atmospheric surface layer (ASL) (Fig. 1). Surface fluxes

measured within the ASL represent the exchange between the surface and the atmosphere (Stull 1988, Rannik 1998). The exchange can be described either by means of a flux ( $F$ ) or the deposition velocity ( $v_d$ ). Their relationship is given by

$$v_d = F / c \quad (2)$$

where  $c$  is the concentration at the measuring height. It is evident from this definition that deposition velocities measured at different sites are not directly comparable unless they have been measured at identical heights. However, for  $O_3$ , for instance, deposition velocity is a better concept than flux because  $v_d$  depends on surface properties that have a major influence on the deposition of  $O_3$  and because the concentration of  $O_3$  varies markedly over a day. On the contrary, for  $CO_2$ , deposition velocity is not an applicable concept because of its high emissions from the surface (Wesely and Hicks 2000).

## 4. Eddy-covariance technique

### 4.1. Basic idea

The basic idea behind eddy covariance (EC) is very simple. Turbulent eddies transport matter and energy vertically within the ABL. The vertical wind speed  $w$  and a scalar such as concentration  $c$  are measured from a few to several tens of metres above a surface, within the ASL (Fig. 2). The measuring frequency of the system is usually about 10 Hz. The measured variables are divided into an average and a fluctuating component (denoted with a bar and a prime, respectively (Seinfeld and Pandis 1998)):

$$\begin{aligned} w &= \bar{w} + w' \\ c &= \bar{c} + c' \end{aligned} \quad (3)$$

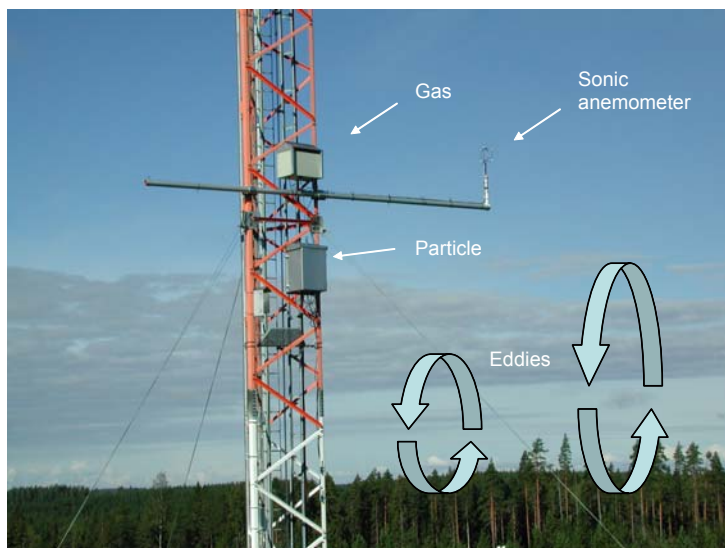


Figure 2. Turbulent eddies transport matter and energy vertically between surface and atmosphere. The eddy-covariance system consists of a sonic anemometer measuring wind speed in 3 dimensions and a gas analyser (gas) or a particle counter (particle) (Photo by P. Hari).

The covariance between the fluctuating components is the eddy flux  $F_{EC}$  of the desired substance:

$$F_{EC} = \overline{w'c'} \quad (4)$$

In forest systems, particularly under conditions of low turbulence, accumulation or depletion of the measured species under the EC measuring system can be considerable, and the change in it can be accounted for with a *storage term*  $F_{st}$  assessed with concentration profile measurements at two or more heights below the EC system:

$$F_{st} = \int_0^{z_m} \frac{\partial c(z)}{\partial t} dz \quad (5)$$

where  $z_m$  is the EC measuring height and  $c$  is the concentration. Although  $F_{st}$  can be large over short-term measurements, it usually averages out to zero over a day or a longer period. Thus the total flux between a forest and the atmosphere is

$$F = F_{EC} + F_{st} \quad (6)$$

#### 4.2. Problems associated with EC

Two major problems in eddy covariance are the imperfect energy balance closure (EBC, see 5.2) and the likely underestimation of night-time respiration ( $\text{CO}_2$ ) fluxes (Aubinet *et al.* 2000, Wilson *et al.* 2002). At most eddy-covariance sites, a positive correlation exists between friction velocity  $u^*$  and night-time  $\text{CO}_2$  flux under conditions of low turbulence (low  $u^*$ ), even if  $\text{CO}_2$  accumulation is taken into account with  $F_{st}$  (Aubinet *et al.* 2000) (Fig. 3). Since respiration as a biological phenomenon should not be dependent on turbulent conditions, this means that the EC measurement most likely underestimates night-time  $\text{CO}_2$  release at low  $u^*$ . Furthermore, during periods with large EBC imbalance, average  $\text{CO}_2$  flux is approximately 15% lower in daytime and 50% lower in night time than during periods with small EBC imbalance, and the magnitude of both problems increases with decreasing  $u^*$  (Wilson *et al.* 2002). This suggests that at least some of the factors behind the two problems are similar and related to the EC technique and to the degree of turbulence rather than to biology.

Possible error sources behind these problems are: 1) systematic errors associated with the mismatch of source areas for fluxes and sensors measuring the other components of the energy balance or  $\text{CO}_2$  storage; 2) systematic instrument biases; 3) neglected sinks (storage of heat above or below soil heat plates or in vegetation, storage of  $\text{CO}_2$  in soil pores, diffusion of  $\text{CO}_2$  into ground water); 4) low and high frequency loss of turbulent fluxes; and 5) horizontal and/or vertical advection of heat, water vapour, and  $\text{CO}_2$  (*e.g.* Wilson *et al.* 2002, Aubinet *et al.* 2003a).

In the case of underestimated respiration, the profile measuring system may not detect all of the accumulated carbon, part of which could, for instance, lie under the lowest

measurement point of the profile system (typically at the height of 2 to 4 m) (Gu *et al.* 2004). In the morning when the daytime mixed ABL starts to form, the accumulated CO<sub>2</sub> may be swept in the air quickly with the morning flush. If all of the accumulated carbon is detected then, long-term carbon balances can be calculated accurately. However, summing up the fluxes measured during the night and during the morning flush most often still leads to too low respiration values compared with the driving variables (Aubinet *et al.* 2000 & 2003b).

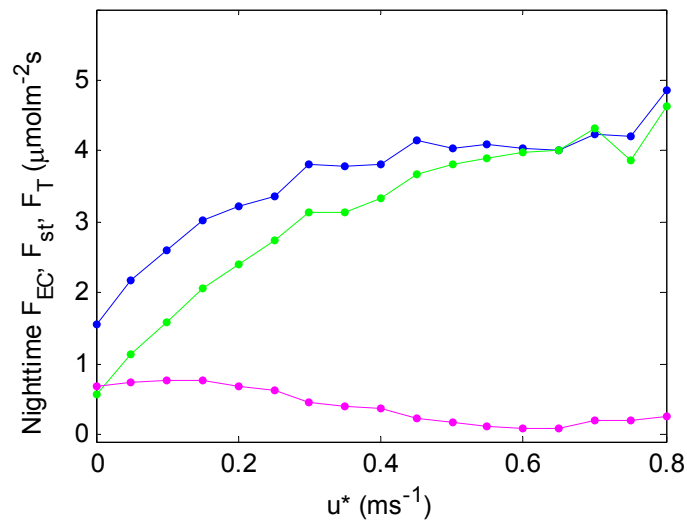


Figure 3. Relationship between  $u^*$  and night-time CO<sub>2</sub> flux (sun elevation angle  $< -3^\circ$ ). Data includes all months of June 1996 - 2002.  $F_{EC}$  = eddy flux (green),  $F_{st}$  = storage flux (change in storage; pink),  $F_T = F_{EC} + F_{st}$  (blue).  $F_{st}$  decreases and  $F_{EC}$  increases with increasing  $u^*$ , but their sum,  $F_T$ , also appears to increase at least at low  $u^*$ .

Most eddy-covariance sites are subject to advection that can result from sloping topographies but also from heterogeneous land use. A slope may lead to the accumulated carbon and water vapour flowing downhill during the night (Fig. 4). As nocturnal evapo-transpiration (latent heat flux) is relatively small, the problem is probably not relevant from the point of view of EBC, but from the point of view of measuring the large nocturnal CO<sub>2</sub> efflux it may be crucial. A solution is to replace measurements made during low turbulence with modelled values. This requires assessing a threshold  $u^*$  value below which the fluxes are modelled and above which the measured fluxes are assumed to be independent of  $u^*$  - a problematic assumption, since changing the subjectively determined threshold appears to affect annual CO<sub>2</sub> balances (Aubinet *et al.* 2000). Furthermore, there is not much evidence that such a threshold exists for the EBC imbalance, either (Wilson *et al.* 2002). Automated, systematic methods for  $u^*$  threshold determination are under development (Gu *et al.* 2004). However, in connection with the modelling method, measuring  $F_{st}$  is vital because otherwise, if the morning flush does include some accumulated carbon, it will be counted twice, once in the modelling and a second time in the measurement (Aubinet *et al.* 2002). Another problem may be that the EC method does not capture the morning flush correctly. During the flush, turbulence is not stationary; however, stationarity is a prerequisite for the EC method and the lack of it may lead to miscalculation of the flux (Foken and Wichura 1996). Moreover, the sonic anemometer used in the measurement typically requires that the angle between the

mean wind direction and the horizontal be less than  $20^\circ$  in order for the equipment itself not to interfere with the air flow and disturb the measurement; in a recent study, however, Gash *et al.* (2003) found that during a one-hour morning flush above a peat bog in the Netherlands, about 60% of the angles were outside  $0$ - $20^\circ$ , mostly between  $-90^\circ$  and  $20^\circ$ . They argued that this phenomenon could contribute to the night-time flux underestimation.

In **Paper III**, we studied the effect of correcting for the night-time stable periods on annual cumulative  $\text{CO}_2$  exchange estimates. We compared two calculation methods: In the first one, we replaced low- $u^*$  measurements with modelled values; in the second we did not. However, both methods took into account  $F_{\text{st}}$ . We found that with the second method the annual net sink was systematically larger, which probably means that the profile measurement did not capture all accumulated  $\text{CO}_2$ . Because the SMEAR II site is a gently sloping hill, it is likely that during stable periods some of the  $\text{CO}_2$  accumulated near the surface, flowed downhill escaping the measuring system (Fig. 3), and was therefore not detected even in the morning.

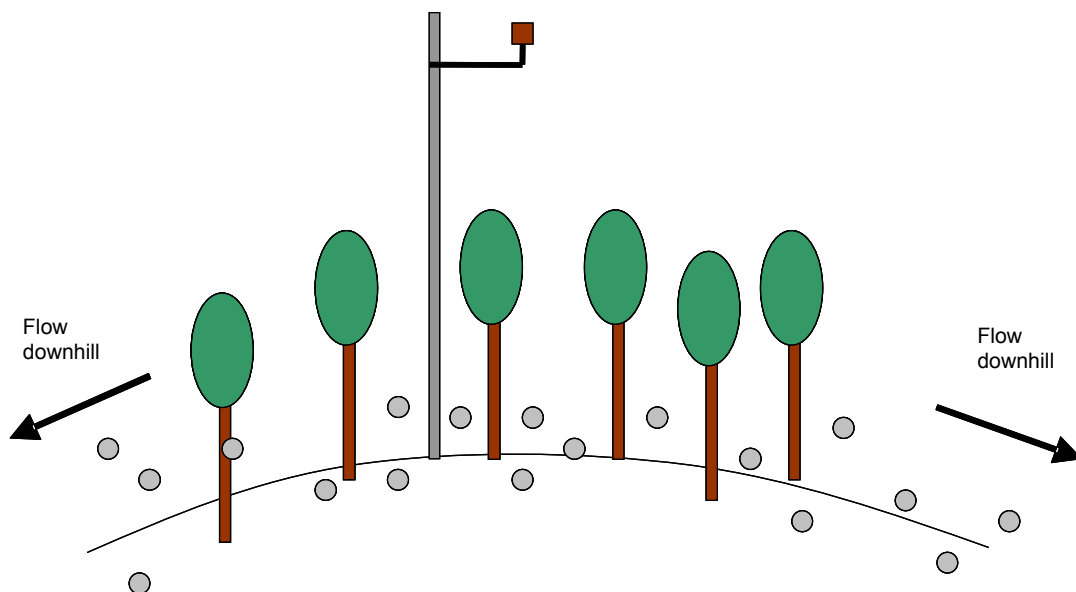


Figure 4. Effect of sloping topography. During stable stratification,  $\text{CO}_2$  emitted from the soil and vegetation (grey circles) accumulates near the surface. If the terrain is not entirely flat, the gas may flow downhill carried by cold air and may not be detected either by the profile measurement system or by the EC measurement system in the morning flush.

The spatial representativeness of the flux is determined by footprint analysis, a tool for locating the source or sink areas upwind from the flux (Markkanen *et al.* 2003). The source area size and upwind distance increase with measurement height and with thermal stability (Aubinet *et al.* 2003a). Because of the latter, flux measurements made in heterogeneous forests may represent completely different ecosystems at night and by day. This must be kept in mind also when comparing flux results from chamber measurements and EC measurements: The chambers always represent a very small,

localised area, whereas the EC method describes a variable area up to several hundreds of metres upwind.

Regardless of the above considerations and caveats, the undisputed benefit of the EC technique is that it is the only method for direct measurement of whole-ecosystem fluxes. This makes it useful for detecting whole-ecosystem responses (**Papers II, IV**), and, to a certain degree, also for calculating annual sums and balances of different substances. Local annual estimates are susceptible to large errors (**Paper III**), but they are useful combined in a larger context within a representative flux measurement network such as FLUXNET.

## 5. Measurements at the SMEAR II field station

**Paper III** describes the measuring systems and the site in detail. Therefore, only the main properties are described here.

### 5.1. Site description

The SMEAR II station is located in a homogeneous Scots pine stand (*Pinus sylvestris* L.), established in 1962 through direct sowing after clear felling and prescribed burning next to the Hyytiälä forest station in southern Finland (61°51'N, 24°17'E, 181 m above sea level). The forest is representative of Finnish forests in general (**Paper III**). The homogeneous fetch in the prevailing wind direction (230°) is 250 m (Vesala *et al.* 1998). The soil is composed of sandy and coarse silty glacial till. The annual mean temperature in 1961—1990 was +2.9 °C and the annual mean precipitation 709 mm. The height of the dominant trees is about 14 m. The all-sided leaf area index (LAI) is approximately 8 m<sup>2</sup>m<sup>-2</sup> (**Paper IV**) and the tree biomass about 68 t ha<sup>-1</sup> (above- and below-ground) (Ilvesniemi and Liu 2001).

Between January and March 2002, an area of 4.33 ha (approximately half of the stand; mainly the southern sector) was manually thinned, and LAI in the thinned area dropped from 8 to 6 (**Paper IV**). At the time of the harvest, the average basal area of the stems at the height of 1.3 m was 24.3 m<sup>2</sup>ha<sup>-1</sup>, and the basal-area-weighted average height was 13.02 m. In the thinning, 6.4 m<sup>2</sup> (= 26%) of the basal area was removed.

### 5.2. Measurements

At the SMEAR II station, EC flux measurements of CO<sub>2</sub>, momentum, and sensible and latent heat have been carried out continuously since April 1996 in a tower (Tower 1) at the height of 23.3 m and occasionally at the height of 46.0 m. Particle fluxes have been measured intermittently from June 1996 until April 1999, and continuous measurement began in June 2000. O<sub>3</sub> measurements have been carried out at the height of 22.0 m in a second measurement tower (Tower 2) since August 2001. The EC fluxes are calculated as 30-min averages of the covariance of vertical wind speed and the scalar in question, such as gas concentration. The measurement system in Tower 1 consists of an ultrasonic 3-D fast-response anemometer (Solent 1012R2) and a fast-response

infrared absorption gas analyser (LI-6262 CO<sub>2</sub>/H<sub>2</sub>O analyser). The CO<sub>2</sub> storage term (the accumulation of CO<sub>2</sub> below the measuring level) is estimated by means of gas gradient measurements at the heights of 4, 8, and 16 m with an infrared absorption analyser (URAS 4). The particle EC system consists of the above-mentioned anemometer at the height of 23 m and a condensational particle counter (CPC) TSI model 3010. The measurement system for O<sub>3</sub> flux in Tower 2 consists of a fast-response acoustic anemometer (Solent HS1199) and a fast response chemiluminescence O<sub>3</sub> analyser (LOZ-3).

The aerosol size distribution measurements in Hyytiälä are performed using a differential mobility particle sizer (DMPS), which has a detection range of 3–600 nm in particle diameter. From the DMPS size distribution plots, the particle formation events (see 5.4) are visually divided into three different classes according to their strength and clarity (**Paper III**).

## 6. Fluxes and ecosystem functioning

Surface fluxes above ecosystems reflect variations in incoming and outgoing energy and in the metabolic activity of vegetation and soil microbes. A prerequisite for interpreting the fluxes is to consider the sources and sinks of each compound in question and to understand the underlying mechanisms and environmental drivers. Figure 5 presents the time series of fluxes of momentum, sensible heat, water vapour/latent heat, carbon dioxide, ozone, and aerosol particles ( $F_m$ ,  $F_H$ ,  $F_w/F_{LE}$ ,  $F_C$ ,  $F_{O_3}$ ,  $F_p$ , respectively) from April 1996 to the end of 2001 measured in Hyytiälä.

In this thesis, the word *ecosystem* is used in a very narrow sense. Basically, it refers to the forest area encompassed by the varying footprint of the eddy-covariance measuring system. The definition includes all vegetation and supposedly excludes all fauna except soil microbes. However, this definition is inaccurate in the case of CO<sub>2</sub> since the total ecosystem respiration does include respiration by animals in addition to that of vegetation. Furthermore, carbon is stored in both vegetation and animal biomass; the former we can estimate with inventory methods, but the latter is an unquantified part of the ecosystem carbon balance.

### 6.1. Carbon dioxide

$F_C$  measured above the forest with the EC system is equal to the *net ecosystem exchange of CO<sub>2</sub>* (NEE). NEE is a sum of both plant *gross primary production* (GPP), driven by photosynthesis, and *total ecosystem respiration* (TER). Photosynthesis fixes carbon from the atmosphere and respiration releases it. In atmospheric science, in which the point of reference is the atmosphere, carbon uptake by photosynthesis is usually denoted negative and release by respiration positive. Part of the carbon-containing sugars produced in photosynthesis as GPP remains in the trees as construction material for new cells and constitutes the growth of each year. This fraction remaining in the tree biomass is called the *net primary production* (NPP) (Fig. 6). The rest of GPP is transported into all living parts of the tree (leaves, branches, stems, roots, and mycorrhizae) and consumed in tree metabolism as *autotrophic respiration* ( $R_a$ ). Microbes in the soil consume dead organic material and thus return the carbon within it back to the atmosphere in *heterotrophic respiration* ( $R_h$ ). NPP is



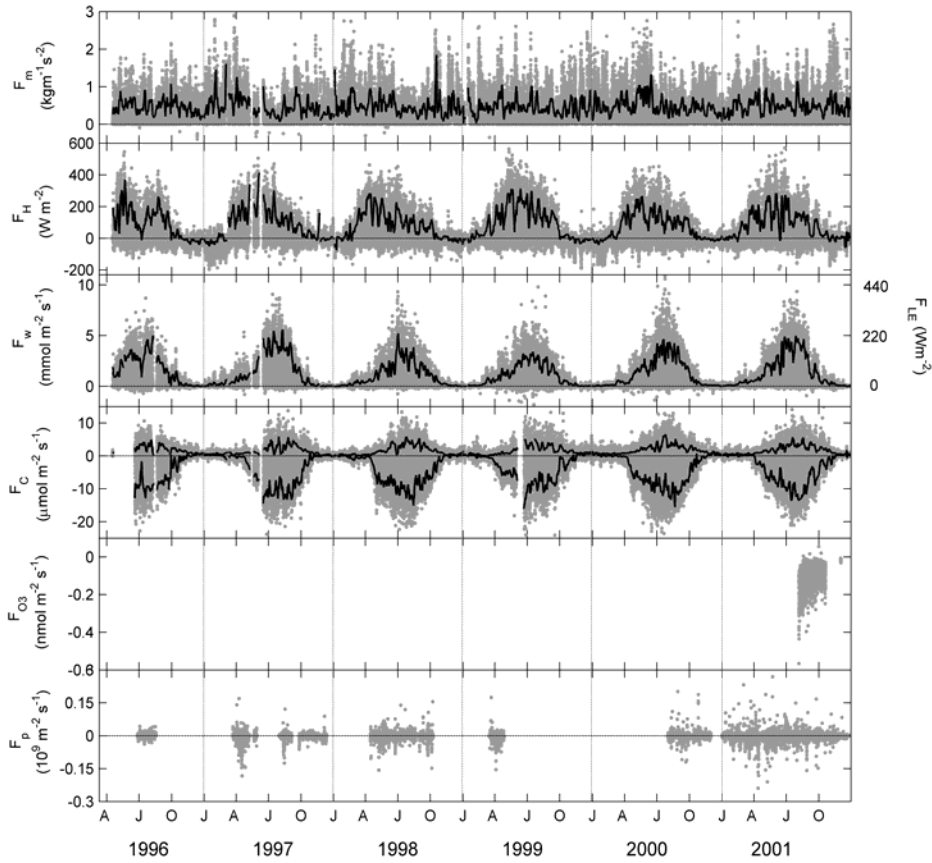


Figure 5. Time series of surface fluxes in Hyytiälä, 1996 – 2001. Grey data points are half-hour averages. Black line in the four top panels is the average maximum day flux (during 11:00–13:00; for  $F_C$  also night flux during 01:00–03:00 (the upper curve)) of the previous 5 days.  $F_m$  = momentum flux,  $F_H$  = sensible heat flux,  $F_w$  = water vapour flux (also presented as  $F_{LE}$  = latent heat flux),  $F_C$  =  $\text{CO}_2$  flux including storage,  $F_{O_3}$  =  $\text{O}_3$  flux, and  $F_p$  = flux of aerosol particles in the size range of 3–600 nm. Sign convention:  $<0$  downwards,  $>0$  upwards. However, for  $F_m > 0$  downwards. From **Paper III** (Fig. 1).

the sum of negative GPP and positive  $R_a$ , and TER the sum of both  $R_h$  and  $R_a$  (Fig. 6). Therefore we can write

$$\text{NEE} = \text{GPP} + \text{TER} = (\text{NPP} - R_a) + (R_a + R_h) \quad (6)$$

In plant ecology, the point of reference is usually the plant or the vegetation, and instead of NEE, plant ecologists use the term net ecosystem production (NEP). The relationship between NEE and NEP is simple:

$$\text{NEE} = -\text{NEP} \quad (7)$$

$\text{CO}_2$  is an *inert* gas that does not take part in chemical reactions in the atmosphere. The diurnal behaviour of NEE is therefore mainly controlled by plant and soil metabolic activity, which, in turn, depend on environmental variables in a non-linear way. The effect of the non-linearity is that long-term average climatic variables are not able to explain NEE except in extreme conditions as in the case of spring night flux or summer drought (**Paper I**). In the boreal zone, photosynthesis occurs in sunlight during the growing season and, to a small extent, also during warm spells in evergreen forests in wintertime (**Paper II**). Usually, however, because the combination of low

temperatures and high irradiance is detrimental to the photosynthetic apparatus, photosynthesis is inhibited in boreal winters (Ottander *et al.* 1995).  $R_h$  occurs in the soil and depends on soil temperature, soil moisture, and the amount of organic substance available for respiration (Kramer and Kozlowski 1979).  $R_a$  occurs both below and above ground and depends similarly on soil temperature and moisture, but also on air temperature, irradiance, plant tissue age and physiological condition, the amount of starch or sugar available for respiration, and, importantly, on current year's photosynthesis (Kramer and Kozlowski 1979, Högberg *et al.* 2001). Root respiration alone has been estimated to comprise 10 to 90% of total soil CO<sub>2</sub> efflux (Lankreijer *et al.* 2003), but it is easy to understand that the relative contributions of  $R_h$  and  $R_a$  to TER vary strongly according to local conditions and time of year. On average, the estimated contribution of soil respiration to TER in Hyytiälä is 50 – 80% (Pumpanen *et al.* 2003 & pers. comm.). At boreal latitudes, overall soil respiration typically diminishes but does not cease in wintertime because the thick snow cover insulates the soil and prevents freezing soil temperatures. Furthermore, the high soil water content results in a large release of latent heat during freezing (Sevanto *et al.* 2004).

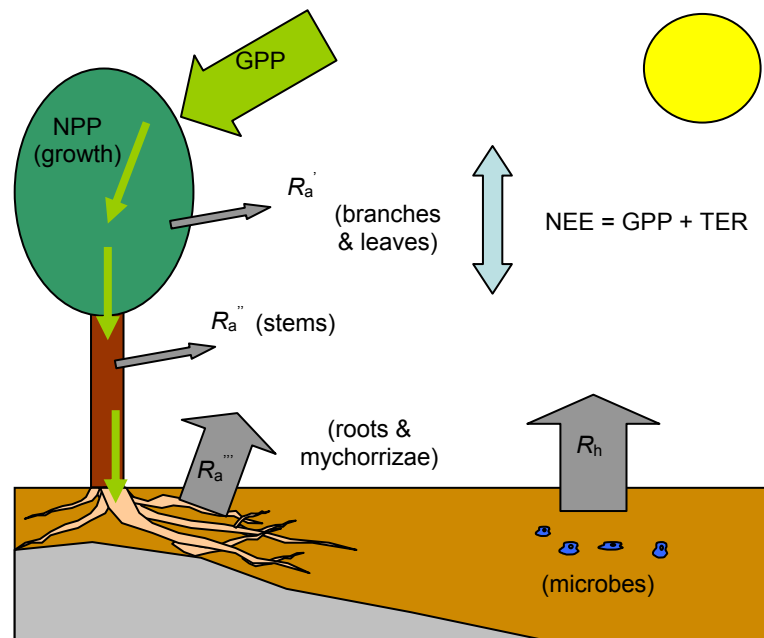


Figure 6. Carbon cycle in a forest ecosystem. Net ecosystem exchange of CO<sub>2</sub> (NEE) is the sum of gross primary production (GPP; negative) and total ecosystem respiration (TER; positive); TER consists of autotrophic respiration ( $R_a$ ; primes denote different sources) and heterotrophic respiration ( $R_h$ ). Net primary production (NPP) is the sum of GPP and  $R_a$ .

On European scale, annual cumulative NEE in different types of forests ranges typically from -700 to +100  $\mu\text{molm}^{-2}$ , net uptake decreasing towards higher latitudes with increasing respiration (Valentini *et al.* 2000). At different sites, cumulative NEE may be rather constant or exhibit great interannual variability, varying by a factor of two or four in the course of a few years (Goulden *et al.* 1996, Lindroth *et al.* 1998, Black *et al.* 2000) depending on changes in photosynthesis and/or respiration (**Paper I**). Many boreal ecosystems are sources of carbon in winter and sinks of carbon during the growing season in summer. The transition from the winter stage to the summer stage is more flexible in coniferous than in deciduous trees because the seasonality of coniferous species is not determined by leaf-out or leaf senescence (Kramer and

Kozłowski 1979). For both evergreen and deciduous forests, the onset of the growing season has been connected with increasing air and soil temperatures and the thawing of the soil in spring (Bergh and Linder 1999, Hollinger *et al.* 1999, Jarvis and Linder 2000). In **Paper II**, we showed that the onset of photosynthesis in the spring in five coniferous forests in Finland, Sweden, and Siberia was triggered by longer-term ( $> 1$  day) average air temperature (Fig. 7). In the autumn, photosynthesis decreases with decreasing temperature, light intensity, and day length (Kramer and Kozłowski 1979). The ending of the growing season is less abrupt in evergreen forests than in deciduous ones; if winters are warm enough, photosynthesis in evergreen stands can continue throughout the year (Pharis *et al.* 1970, Kramer and Kozłowski 1979, Schaberg *et al.* 1995, Hansen *et al.* 1996, Anthoni *et al.* 1999, Berbigier *et al.* 2001).

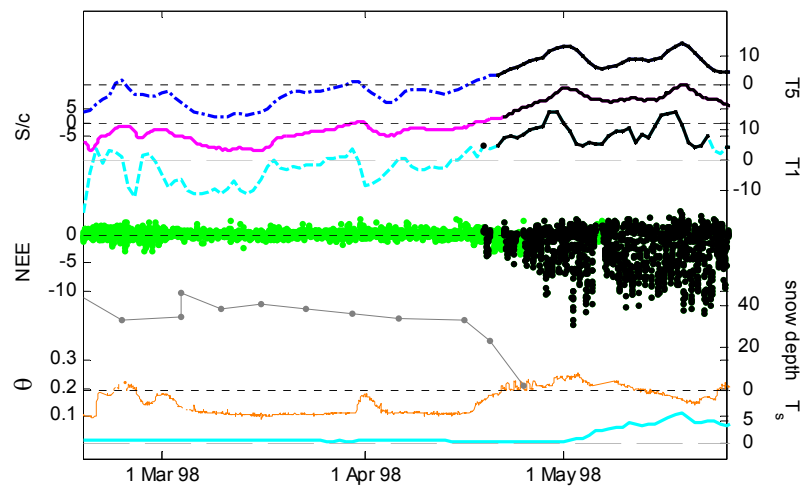


Figure 7. Spring recovery in Hyytiälä, 1998. From top to bottom: dark blue = 5-day average air temperature  $T_5$  ( $^{\circ}\text{C}$ ), pink = stage of development of the ecosystem  $S/c$  (depends on air temperature), light blue = 1-day average air temperature  $T_1$  ( $^{\circ}\text{C}$ ), green = NEE ( $\mu\text{molm}^{-2}\text{s}^{-1}$ ), grey = snow depth (cm), orange = soil water content in humus  $\theta$  ( $\text{m}^3\text{m}^{-3}$ ), light blue = soil temperature at 5 cm  $T_{s5}$  ( $^{\circ}\text{C}$ ). The dashed lines from top to bottom indicate zero levels. The temperature-related variables ( $T_1$ ,  $T_5$ ,  $S/c$ ) are used for *predicting* the spring recovery. After they have exceeded an empirical threshold that should trigger the recovery, the curves are coloured black. After the *observed* spring recovery, the NEE curve is coloured black. The coincidence of observed and predicted spring recovery is evident. From **Paper II** (Fig. 5).

In principle, the EC technique allows also the separation of the measured NEE into component fluxes, TER and GPP, after daytime TER has been estimated with, for instance, a temperature regression function on the basis of night-time measurements and subtracted from GPP. This approach is appealing and widely used, but at the moment, the reliability of such calculated fluxes is poor because 1) extrapolating the temperature regression from low night-time temperatures to high daytime temperatures may be arbitrary; 2) using a simple temperature regression based on, at best, air temperature and soil temperature measured at one single depth in the soil is altogether unphysical: Respiration in the soil occurs at all depths and depends on the whole soil temperature profile, which, in turn, depends on air temperature with a varying time lag. For instance, already at the depth of 5 cm in the A-profile (the uppermost mineral soil layer just below the humus) the time lag between soil and air temperature is 8 – 12 h depending on soil composition and the depth of the humus layer; 3) finally, leaf dark respiration is expected to be inhibited in light, possibly by about 50% on average, but

the degree of inhibition changes with leaf age and light intensity (Villar *et al.* 1994 & 1995), and as a result, daytime TER cannot be accurately estimated on the basis of night-time measurements.

## 6.2. Water vapour (latent heat)

Water vapour has two important functions in a forest-atmosphere system. First, it is an integral component of the ecosystem water balance: Water enters the terrestrial ecosystem as precipitation, is stored in the soil and taken up by vegetation, and returns to the atmosphere by *evaporation* and *transpiration*. At the same time, along with  $F_H$ ,  $F_w$  (or, equivalently,  $F_{LE}$ ) is a major contributor to the ecosystem's energy balance (Shaw and Finnigan 2002).

$F_w$  measured above a forest is equivalent to the *evapo-transpiration* of the ecosystem, the sum of transpiration by vegetation and evaporation from surfaces. They both depend on temperature, radiation, soil moisture, and vapour pressure deficit (VPD), but, unlike evaporation, transpiration is biologically controlled. In order to photosynthesize, the plant has to open stomata and let atmospheric CO<sub>2</sub> diffuse in. However, at the same time water vapour diffuses out. As a result, the plant monitors the amount of transpirable water and whether or not the conditions are suitable for photosynthesis and controls the stomatal aperture accordingly (Hari *et al.* 1986, Taiz and Zeiger 1998). Biological factors are very important also for total ecosystem water cycling. For example, moderate foliage removal does not change the total ecosystem evapo-transpiration rate probably because the canopy and ground vegetation compensate for the foliage removal by redistributing the sources very efficiently (Fig. 8, **Paper IV**). With less competition, trees and understorey plants get a larger portion of the available soil water and can afford to transpire with a higher rate. Furthermore, the understorey evapo-transpiration increases with increasing PAR reaching the forest floor.

The relationship among net radiation  $R_n$  and the other energy balance components can be expressed as

$$R_n + F_G + F_M = F_H + F_{LE}, \quad (8)$$

where  $F_G$  is the energy stored in the soil or canopy,  $F_M$  is the energy associated with biological processes such as photosynthesis and respiration,  $F_H$  is the flux of sensible heat, and  $F_{LE}$  the flux of latent heat ( $L$ =latent heat of vaporization and  $E$ =flux of water vapour). For plant surfaces,  $F_M$  is so small compared with the other components that it is usually neglected (Snyder and Paw U 2002). The left-hand side of Eq. (8) represents the energy available for evapo-transpiration and  $F_H$ . Plotting it against  $F_H + F_{LE}$  usually reveals a deficit in energy balance closure at eddy-covariance measurement sites (Wilson *et al.* 2002). The partitioning of energy between  $F_H$  and  $F_{LE}$  is significant: In the daytime when the convective mixed ABL prevails, the main source of turbulence is  $F_H$ . In boreal forests, the sensible heat flux is often large and, consequently, ABL grows deeper than in temperate forests where latent heat fluxes dominate over sensible heat (Baldocchi and Vogel 1996).

The relative contributions of evaporation and transpiration to  $F_w$  vary: On dry summer and autumn days, the diurnal cycle of  $F_w$  above a boreal forest is mostly explained by plant transpiration (*e.g.* Sevanto *et al.* 2001), whereas after precipitation events and in winter and spring, evaporation from wet ground or snow becomes significant.  $F_w$  is mainly positive throughout the year, but close to zero during November—February (Fig. 5). In Hyytiälä,  $F_w$  reaches maximum values of 7 to 10  $\text{mmolm}^{-2}\text{s}^{-1}$  in June (corresponding to  $F_{LE}$  of 300 to 440  $\text{Wm}^{-2}$ ).

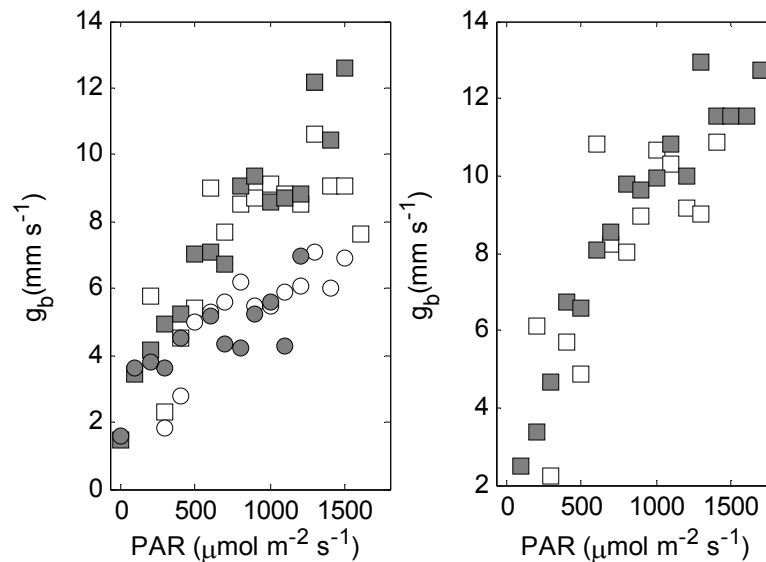


Figure 8. Effect of thinning on the ecosystem bulk conductance  $g_b$ , calculated with two methods: by normalizing 1) measured  $F_w$  by VPD and 2)  $F_{O_3}$  by ozone concentration ( $c_{O_3}$ ). The figure shows a comparison of  $g_b$  between thinned and intact areas in June–September at RH of 40 – 70% as a function of PAR. Each data point represents half-hour measurements averaged over a PAR interval of 100  $\mu\text{molm}^{-2}\text{s}^{-1}$ . Left:  $F_w/\text{VPD}$  after the thinning in 2002 for the intact areas (grey squares) and for the thinned areas (white squares);  $F_{O_3}/c_{O_3}$  in 2002 for the intact areas (grey circles) and for the thinned areas (white circles). Right:  $F_w/\text{VPD}$  for the whole south-east sector in 1996–2001 before the thinning (grey squares) and in 2002 after the thinning (white squares). From **Paper IV** (Fig. 3).

### 6.3. Ozone

Ozone is a key species in atmospheric chemistry and a greenhouse gas that is potentially toxic to living organisms. The main sources of tropospheric  $O_3$  are local photochemical production in the presence of volatile organic compounds (VOCs), nitric oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ), and sunlight, and intrusion of  $O_3$ -rich air from the stratosphere (*e.g.* Jenkin and Clemitshaw 2000).  $O_3$  is *reactive*: It is destroyed upon contact with surfaces as well as in gas-phase reactions. In a forest canopy,  $O_3$  sinks consist of stomatal uptake, deposition on non-stomatal surfaces and destruction in gas-phase chemical reactions. Reactions with terpenoids can lead to the formation of secondary aerosols (see 6.4).

The main controlling factor for  $O_3$  deposition during the growing season is the surface uptake resistance which, in daytime, is mainly considered to be stomatal. This partly

explains why O<sub>3</sub> deposition appears to be insensitive to small disturbances such as moderate thinning (Fig. 8, **Paper IV**): When plants transpire with a higher rate (although VPD remains constant), their stomatal aperture is larger which enhances O<sub>3</sub> deposition on individual plants. The toxicity of O<sub>3</sub> to plants depends not only on episodes of high O<sub>3</sub> concentrations but also on accumulated doses of O<sub>3</sub> flux. This is important in boreal forests where water stress is rare and high stomatal conductances are frequent (Simpson *et al.* 2002). However, Kurpius *et al.* (2003) showed that within-canopy chemistry may sometimes be the dominant daytime O<sub>3</sub> loss process, accounting for as much as 55% of the total O<sub>3</sub> sink in summer and about 15% in winter. In their study, the chemical sink showed a temperature dependence which could be explained by temperature-enhanced emissions of terpenoids from vegetation.

The diurnal range of  $v_{d,O_3}$  is about 2–7 mms<sup>-1</sup> in Hyytiälä, which is similar to that at other coniferous sites in Finland and Denmark (Tuovinen *et al.* 2001, Pilegaard *et al.* 1995). Chemical reactions with NO<sub>x</sub> and terpenoids destroy O<sub>3</sub> during night time after the photochemical production has ceased. Even though the temperature-related emissions of monoterpenes from the vegetation decrease at night (Rinne *et al.* 2000, Kulmala *et al.* 2000a, Hakola *et al.* 2000), their ambient concentrations increase because monoterpenes accumulate in the canopy during nights with low mixing and a shallow ABL (Rinne *et al.* 2000). Additionally, the lack of turbulence at night prevents the deposited or chemically destroyed ozone near the surface from being replaced by more ozone from the higher air masses. The ensuing O<sub>3</sub> sink near the surface is sustained if the characteristic times of O<sub>3</sub> chemistry are fast compared with those of the low turbulent transport. This is indeed the case in Hyytiälä according to the results of Keronen *et al.* (2003), which reveal the existence of a clear vertical profile in  $c_{O_3}$  with concentrations decreasing towards the surface. The formation of dew on the surface of needles and other vegetation in the morning hours could also provide an additional sink inside the canopy (Lamaud *et al.* 2002) since in Hyytiälä, the total night-time deposition increases also with increasing RH (Altimir *et al.* 2004, Altimir *et al.* in prep.).

Towards winter, O<sub>3</sub> deposition decreases because the production and therefore the concentrations of O<sub>3</sub> are reduced and the stomatal uptake of the vegetation diminishes. Moreover, the resistances of a coniferous forest canopy and ground to O<sub>3</sub> deposition are about 1.5 and 17.5 times higher with snow cover than without it, respectively (Seinfeld and Pandis 1998). Even upward  $F_{O_3}$  from the surface of snow has been observed. Zeller (2000) postulated that O<sub>3</sub> may be stored in the snow or in tree stands. Also, UV radiation may photolyse NO<sub>3</sub> in snow and result in surface emission of NO<sub>x</sub> which leads to the production and subsequent upward transport of O<sub>3</sub> (Dibb *et al.* 2002).

#### **6.4. Aerosol particles and terpenoids**

An aerosol consists of liquid or solid particles suspended in air. Diameters of *fine particles* range typically from a few nanometres up to 1 µm; *coarse particles* are larger. The atmosphere always contains a variable amount of aerosol particles, even up to 10<sup>7</sup> to 10<sup>8</sup> cm<sup>-3</sup> (Seinfeld and Pandis 1998). Particles enter the atmosphere by emissions from local and remote sources (primary particles) and by gas-to-particle conversion including nucleation and subsequent condensational growth (secondary particles) which depend in a highly non-linear fashion on the concentrations of the

nucleating and condensing vapours, temperature, and relative humidity (Kulmala *et al.* 2000b). Particles are removed from the atmosphere by coagulation (small particles adhering on the surface of larger ones) and deposition. In this thesis, the term *new-particle formation event* or just *formation event* refers to the sudden appearance and subsequent growth of 3-nm particles which are the smallest our measuring system can detect. *Nucleation* refers to the forming of a cluster of molecules that is thermodynamically stable, that is, that does not evaporate away immediately. The nucleated clusters may then grow by condensation to detectable sizes and initiate a formation event. Because in this thesis we consider nucleation only in connection with new-particle formation events, we sometimes use the familiar but imprecise expression *nucleation event*. *Formation* in general refers to either nucleation or condensational growth or to both.

The size distribution of an aerosol measured at a field station changes constantly. Air masses coming from clean or polluted areas carry particles that are always subject to the dynamic processes of condensation and evaporation of organic and inorganic vapours onto and from their surface. New-particle formation events have been observed frequently in the continental boundary layer in many parts of the world (Kulmala *et al.* 2004): from sub-arctic Lapland to the remote boreal forest in southern Finland (Mäkelä *et al.* 1997), and from central Europe (Birmili and Wiedensohler 2000) to rural United Kingdom (Coe *et al.* 2000). The most probable formation mechanism is the ternary nucleation process  $\text{H}_2\text{SO}_4\text{-H}_2\text{O-NH}_3$  (Kulmala *et al.* 2000b). However, evidence suggests that also biogenic formation occurs in the form of condensation of organic vapours on the surface of the new particles (Kulmala *et al.* 2001). Nucleation appears to take place at high latitudes only in cold-air advection of Arctic or polar air masses, with low cloudiness, with close connection to the onset of strong turbulence, convection, and entrainment in the morning. The small particles then deposit downwards and grow as a result of organic-vapour condensation (Kulmala *et al.* 2000a). Terpenoids that are emitted by vegetation are likely to lead to aerosol formation. They are biogenic volatile organic compounds (BVOC's) that constitute the largest class of *secondary metabolites*, side products of photosynthesis (Taiz and Zeiger 1998). They do not appear to have any direct function in plant growth and development, but we know that some of them attract pollinators and defend plants against herbivores. In conifers, monoterpenes ( $\text{C}_{10}$ ) accumulate in resin ducts in the needles, twigs, and trunk, the dominant monoterpenes being  $\alpha$ - and  $\beta$ -pinene, limonene, and myrcene. In some species, sesquiterpenes ( $\text{C}_{15}$ ) are found in glandular hairs on the plant surface (Taiz and Zeiger 1998). The volatility of these terpenoids themselves is too high for direct condensation on the surface of existing aerosol particles, but through reactions with atmospheric oxidants  $\text{O}_3$ ,  $\text{OH}$ , and  $\text{NO}_3$  (Atkinson 2000), they form products with high molecular weight and low volatility that readily take part in gas-to-particle conversion (Calogirou *et al.* 1999, Atkinson 2000, Raes *et al.* 2000, Jenkin and Clemitshaw 2000). Pinic acid, for instance, a degradation product of both  $\alpha$ - and  $\beta$ -pinene, is probably very involatile and is likely to contribute to secondary aerosol formation by nucleation or condensation (Raes *et al.* 2000, Jenkin and Clemitshaw 2000, Janson *et al.* 2001).  $\alpha$ -pinene is a major constituent in monoterpene fluxes in Hyytiälä (Rinne *et al.* 2000, Janson *et al.* 2001). Kurpius *et al.* (2003) suggest that sesquiterpenes, which react with  $\text{O}_3$  much faster than monoterpenes but whose emissions have a similar dependence on temperature, could be an important contributor to aerosol formation. However, because of their short lifetimes, they are more difficult to measure than monoterpenes.

In addition to chemical reactions, physical, meteorological, and biological factors enter into the life cycles of *multireactive* aerosol particles. Because aerosols reduce the amount of solar radiation reaching the surface, they influence  $F_H$  and  $F_w$  and the evolution of the ABL (Yu *et al.* 2002). There is also some indication that the deposition of hygroscopic salt particles on leaf surfaces may have an effect on stomatal water transport processes (Burkhardt *et al.* 2001). Many more important links and feedbacks among forests, aerosols, and climate are possible (**Paper V**, Fig. 9).

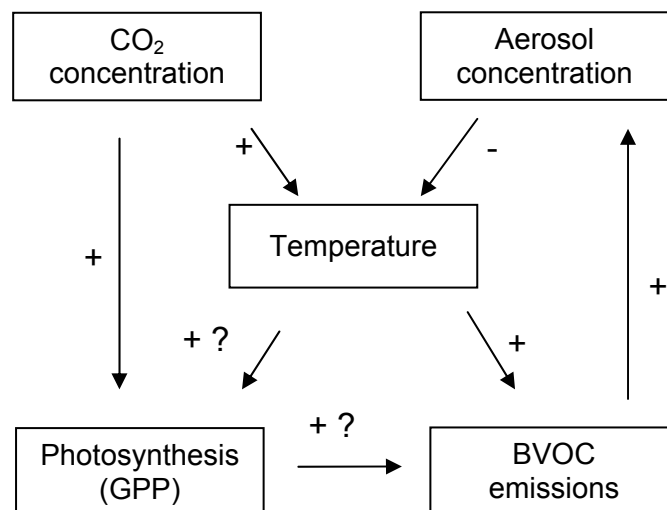


Fig. 9. Schematic figure of coupling of atmospheric  $\text{CO}_2$  concentration, assimilation of carbon by vegetation (gross primary production GPP), emission of biogenic volatile organic compounds (BVOCs), and aerosol particle concentration with atmospheric temperature. Increased  $\text{CO}_2$  concentration will increase temperature (+) and vegetation productivity (+). Increased temperature will enhance BVOC emissions (+) and probably also plant productivity (+?). Increased vegetation productivity may enhance BVOC emissions (+?). Increased BVOC emissions will enhance aerosol formation and growth and therefore also enhance CCN concentrations (+). Enhanced aerosol and CCN concentrations will decrease temperature (-) because of increased reflection of sunlight from low clouds back to space. This results also in the increase of diffuse radiation, which has a positive influence on photosynthesis (Gu *et al.* 2003) (From **Paper V**, Fig. 1).

Measuring aerosol fluxes reliably is difficult. Considering the fact that our measuring devices are able to measure particles only within their lower and upper detection limits and even then the aerosol is subject to losses in the tubes, it is clear that we always observe only a part of the existing aerosol population. This is especially significant for deposition velocity calculations based on eddy-covariance measurement that simply counts the number of particles without giving information on their sizes: For a polydisperse aerosol, total  $v_{d,p}$  is a function of  $v_{d,p,s}$  of all aerosol constituents and highly dependent on the individual characteristics of different particles. Consequently,  $v_{d,p}$  is not very easy to interpret for an integrated size distribution. The average  $v_{d,p}$  towards the forest is greater on nucleation event days than on other days most likely because the small nucleation-mode particles have greater deposition velocities than the larger accumulation-mode particles that prevail on non-event days (**Paper III**).



## 7. Review of publications

The papers comprising this thesis deal with three different, partly overlapping aspects. Firstly, with carbon cycles, their variability and timing (**Papers I & II**); secondly, the effect of climatic factors and human-induced disturbance on the fluxes of inert, reactive, and multireactive substances (**Papers I, II, III, and IV**); and thirdly, with interactions among forests, aerosol dynamics, and climate (**Paper V**). All the papers are based on measurements at the SMEAR II forest station in Hyytiälä, southern Finland. In **Paper II**, there are data from four additional boreal forest sites in Sweden, Finland, and Central Siberia.

**Paper I:** We studied the interannual variability of NEE and its connection with cumulative or average climatic variables during five growing seasons. The forest was a sink of carbon with little interannual variability. The average climatic variables explained the variability of the seasonal or growing-season cumulative NEE only partly, suggesting firstly that the influence of short-term fluctuations in environmental variables is important and secondly that ecosystem responses to those variables are not linear. We also found that both the daytime and the night-time CO<sub>2</sub> fluxes contributed markedly to the variability in carbon exchange, indicating that photosynthesis and respiration have an equally important influence on NEE.

**Paper II:** We investigated the environmental cues initiating the spring recovery of boreal coniferous forest ecosystems under field conditions at altogether five field stations located in northern and southern Finland, northern and southern Sweden, and central Siberia. The within- and intersite variability of spring recovery was large, 30–60 days. Of the different climate variables examined, air temperature emerged as the best predictor for the recovery in spring. We also found that ‘soil thaw’, defined as the time when near-surface soil temperature rapidly increases above 0 °C, was not a useful criterion for spring recovery. At four sites, we were able to quantitatively determine a threshold for air-temperature-related variables, the exceeding of which was required for the recovery. However, the threshold values varied from site to site, perhaps reflecting genetic differences among the stands or climate-induced differences in the physiological state of trees in late winter/early spring.

**Paper III:** We analysed the annual and diurnal patterns of surface fluxes measured with EC in Hyytiälä, 1996-2001. We also presented the annual cumulative NEE calculated by two different methods and focused on comparing the fluxes and deposition velocities of CO<sub>2</sub>, O<sub>3</sub>, and aerosol particles. These are atmospheric constituents with very different characteristics, the first being an inert and the second a reactive trace gas, and the multireactive particles taking part in chemical, physical, and biological processes. We found that at the diurnal scale, the flux of inert CO<sub>2</sub> followed climatic factors such as radiation and soil and air temperature very closely, whereas the flux and the deposition velocity of reactive O<sub>3</sub> depended on radiation in daytime but also varied at night probably because of sink reactions. The flux of multireactive aerosol particles showed no clear connection with any climatic variable. Finally, we showed that particle deposition velocity was higher during new-particle formation events most likely because of the relatively larger amount of nucleation-mode particles.

**Paper IV:** We studied the effect of human-induced disturbance on forest-atmosphere exchange of gases and particles. We found that thinning decreased the deposition velocities of fine particles as expected but did not reduce the carbon sink, water vapour flux, or ozone deposition. The insensitivity of trace gas fluxes to thinning is in contrast to present knowledge, and we suggested that the redistribution of sources and sinks within the ecosystem was comprehensively able to compensate for the lower foliage area. The result has a major importance in the debate regarding the effects of management on forest-atmosphere interactions.

**Paper V:** We presented a hypothesis based on the importance of terpenoids as a link among climate, aerosol dynamics, and CO<sub>2</sub>-driven forest metabolism, emphasising the significance of forests with respect to climate change. Forests, in addition to being sinks of the greenhouse gas CO<sub>2</sub>, also act as sources of climate-cooling aerosol particles. For both of these reasons, increased forest growth leads to the slowing down of global warming. For more quantitative estimations, laboratory experiments, global climate modelling, and extensive international measurement campaigns are necessary.

## 8. Conclusions

In terrestrial surface flux studies, particular attention must be paid to the micrometeorological constraints of measurement techniques: Periods of stable stratification and the instationarity of turbulence are important sources of error; neglecting them may lead to misinterpretation of data. In this work, we used the EC method to study the interactions between boreal forests and the atmosphere from various aspects and attempted to take into account typical error sources such as night-time periods of low turbulence.

The forest in Hyytiälä was a sink of carbon, assimilating about 200 g(C)m<sup>-2</sup>yr<sup>-1</sup> on average with a variation of about 80 g(C)m<sup>-2</sup>yr<sup>-1</sup> or less (**Papers I & III**). Although the flux of inert CO<sub>2</sub> followed climatic factors such as radiation and soil and air temperature very closely at the diurnal scale, climatic variables averaged over seasons explained the seasonal variability of NEE only partly (**Paper I**). This suggests firstly that the influence of short-term fluctuations in environmental variables is important and secondly that ecosystem responses to those variables are not linear. We also found that both the daytime and the night-time CO<sub>2</sub> fluxes contributed markedly to the variability in carbon exchange, indicating that photosynthesis and respiration have an equally important influence on NEE (**Paper I**). The recovery of photosynthesis in spring had the best connection with air temperature and not with soil thawing (**Paper II**). We studied five boreal coniferous sites, and at four of them we were able to determine a threshold for air-temperature-related variables, the exceeding of which was required for the recovery. Our results suggest that higher spring temperatures will most likely lead to an earlier spring recovery of photosynthesis and therefore to longer growing seasons in some parts of the boreal zone. On the other hand, before soil temperatures can rise significantly above zero in spring, snow must melt completely. In addition, at sites where the soil freezes in winter, the consumption and release of latent heat in melting and freezing of water in the soil will maintain the soil temperatures close to zero until all the ice has melted (*zero curtain*). Therefore, the effect of rising air temperatures on soil warming and, consequently, soil respiration

appears to be more complicated and less direct than the effect on growing season length.

Climatic factors had a clear seasonal influence on most surface fluxes. This is because cloudiness and time of year affected the amount of incoming energy; furthermore, radiation, temperature, and precipitation influenced the metabolic activity of the ecosystem (**Papers I & III**). They also had a direct effect on the diurnal behaviour of  $F_C$ ,  $F_w$ , and  $v_{d,O_3}$  but no observable effect on  $v_{d,p}$ . On the other hand, analysing particle and trace gas fluxes before and after a thinning in which 25% of the tree stems were removed showed that  $v_{d,p}$  was directly affected by the physical amount of foliage (**Paper IV**). Interestingly,  $F_C$ ,  $F_{wv}$ , and  $v_{d,O_3}$  were insensitive to the reduction in foliage and to the significant increase in light penetration within the canopy and on the ground. Our results suggest that the redistribution of trace-gas sources and sinks within the ecosystem was comprehensively able to compensate for the lower foliage area. Furthermore, we showed that although the flux of multireactive aerosol particles showed no clear connection with any climatic variable,  $v_{d,p}$  was higher during new-particle formation events most likely because of the relatively larger amount of small, nucleation-mode particles (**Paper III**). Finally, terpenoids emerged as a link among climate, aerosol dynamics, and forest metabolism (**Paper V**). Forests, in addition to being sinks of the greenhouse gas  $CO_2$ , also act as sources of climate-cooling aerosol particles. For both of these reasons, increased vegetation growth potentially leads to the slowing down of global warming.

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## Glossary

ABL	The atmospheric boundary layer = the lowest part of the troposphere. Thickness ranges from hundreds of metres (night) to a couple of kilometres (day).
ASL	The atmospheric surface layer = the lowest 10% of the ABL. In this layer, the surface fluxes are approximately constant (above vegetation).
Boreal	Northern.
BVOC	Biogenic volatile organic compound.
Coniferous	Refers to trees with needles.
Deciduous	Refers to trees which shed their leaves annually.
DMPS	Differential mobility particle sizer. Measures the size distribution of aerosol particles. Detection range about 3 – 600 nm in particle diameter.
Dry deposition	Removal of gases and particles from the atmosphere onto surfaces in dry conditions.
EC, eddy covariance	A direct flux measurement method based on the covariance of vertical wind speed and (typically) gas/particle concentration. Used within the ASL to measure the net exchange of a substance between an ecosystem and the atmosphere.
Eddy	A swirl of turbulent air in the ABL. Size ranges from millimetres to several hundreds of metres.
Friction velocity	A measure of atmospheric turbulence.
GPP	Gross primary production. The gross amount of carbon fixed by vegetation as a direct result of photosynthesis. Part of the carbon will be lost in autotrophic respiration, and the fraction remaining in vegetation is NPP. Denoted with a negative sign.
NEE	Net ecosystem exchange of carbon. The net amount of carbon transferred from the atmosphere to an ecosystem or vice versa as a result of photosynthesis and total ecosystem respiration: $NEE = GPP + TER$ . Equals $-NEP$ .
NEP	Net ecosystem production, equals $-NEE$ . The net amount of carbon acquired by an ecosystem as the difference between gain by photosynthesis and loss by total ecosystem respiration.

New-particle formation event	Sudden appearance of 3-nm particles (smallest size detectable with a DMPS system) and their subsequent growth.
NPP	Net primary production. The net amount of carbon fixed by vegetation as the difference between gain by photosynthesis and loss by autotrophic respiration. Equals the increase in plant biomass = growth.
Nucleation	Formation of a cluster of molecules that is thermodynamically stable (does not evaporate away immediately). If the clusters grow to the diameter of 3 nm, they may be detected and constitute a new-particle formation event.
PAR	Photosynthetically active radiation. Includes those wavelengths of sunlight that plants use for photosynthesis.
Precipitation	Rainfall.
$R_a$	Autotrophic respiration = respiration by the vegetation. Plants use part of the carbon acquired in photosynthesis for metabolism: maintenance of old tissue and synthesis of new.
$R_h$	Heterotrophic respiration = respiration by micro-organisms in the soil. Bacteria and fungi decompose organic matter in the soil and release $\text{CO}_2$ in the process.
RH	Relative humidity.
Stoma (pl. stomata)	Small apertures on the surface of leaves/needles through which $\text{CO}_2$ enters the plant and water vapour diffuses out.
Stomatal control	The ability of plants to enlarge/reduce stomatal apertures depending on environmental conditions.
Surface flux	Flux of energy or gas/particles between the atmosphere and the Earth's surface.
TER	Total ecosystem respiration, $R_a + R_h$ . Denoted with a positive sign.
Terpenoids	A class of biogenic volatile organic compounds (BVOC's), side products of photosynthesis.
Transpiration	Loss of water vapour through stomata.
$u^*$	Friction velocity. A measure of atmospheric turbulence.
VPD	Water vapour pressure deficit. The difference between the actual RH (< 100%) and 100%. Driving force for plant transpiration.