Long-term measurements of surface fluxes above a Scots pine forest in Hyytiälä, southern Finland, 1996–2001

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We present nearly six years of measurements of surface fluxes of momentum, sensible heat, water vapour, carbon dioxide and aerosol particles, along with a three-month time series of ozone flux, measured by eddy covariance above a Scots pine forest in southern Finland from April 1996 to December 2001. The results show marked seasonal and diurnal variation and a significant effect of local and remote anthropogenic pollution on CO_2 and particle data. The flux of inert CO_2 followed environmental factors through plant and soil-microbe metabolism very closely. The flux of reactive O_3 depended on the overlapping influences of environmental parameters driving photosynthesis and of the available amount of reaction partners. The flux of multireactive aerosol particles showed no clear connection with any environmental variable. On new-particle-formation days, the average deposition velocity of particles was, however, greater than usual because small nucleation-mode particles have greater deposition velocities than do larger accumulation-mode particles.

Introduction

The biosphere and the atmosphere form a coupled, complex, non-linear system, the components of which interact by exchanging momentum, heat, gases and particles. The vertical flux of momentum (F_m) is a measure of the friction that the surface inflicts on air masses moving above it. This friction effect is confined to the atmospheric boundary layer (ABL), the dynamics of which affect all surface fluxes. In the deep daytime boundary layer, concentrations of gases and particles are mixed effectively. At night, the boundary layer is shallow with mixing less effec-

tive. Consequently, the net emission or deposition of gases and particles exerts 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 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 species in the morning is especially important in boreal coniferous forests where the ABL often grows markedly higher than, for instance, in temperate forests (Baldocchi and Vogel 1996).

Sensible heat and water vapour (latent heat) fluxes (F_H, $F_w(F_{LE})$) are major contributors to the ecosystem energy balance, the key to the nature and development of the ABL (Shaw and Finnigan 2002). F_w above a forest is equivalent to the *evapo-transpiration* of the site, that is, the sum of *transpiration* by vegetation and *evaporation* from the surfaces. Both of these depend on temperature, radiation, soil moisture and vapour pressure deficit (VPD), but, unlike evaporation, transpiration is biologically controlled. 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 attributable mostly to plant transpiration (Sevanto *et al.* 2001), whereas after precipitation events and in winter and spring, evaporation from wet ground or snow becomes significant.

Excluding the effect of local anthropogenic pollution, the flux of CO_2 (F_C) above a forest is equal to the net ecosystem exchange of CO_2 (NEE), a sum of both plant gross primary production (GPP) driven by photosynthesis, and total ecosystem respiration (TER). Carbon dioxide (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. In the boreal zone, photosynthesis occurs in sunlight during the growing season and, to a very small extent, also during warm spells in evergreen forests in wintertime. Usually, however, it is inhibited in boreal winters (Ottander *et al.* 1995). Release of CO_2 results from respiration that takes place in all living plant cells, including the leaves, branches, stems, roots and mychorrizae; respiration by microorganisms within the soil is also important. The relative contribution of these constituents changes in the course of the year according to soil and air temperature, plant tissue age and physiological condition, moisture conditions and the amount of respirable starch or sugar (Kramer and Kozlowski 1979, Högberg *et al.* 2001). At boreal latitudes, overall soil respiration typically diminishes but continues in wintertime, because the thick snow cover insulates the soil and prevents freezing soil temperatures.

Tropospheric concentrations of ozone (O_3), a greenhouse gas that is potentially toxic to living organisms, are globally increasing (Volz and Kley 1988). Stomatal uptake is an important sink of O_3, and the toxicity of O_3 to plants depends not only on episodes of high O_3 concentrations leading to large O_3 fluxes (F_{O3}) but also on accumulated doses. Although boreal forests do not experience the highest ambient concentrations, they rarely experience water stress and are therefore able to sustain high stomatal conductances that can lead to high cumulative doses (Simpson *et al.* 2002). Tropospheric O_3 concentrations depend mainly on a regional photochemical production in the presence of volatile organic compounds (VOCs), nitric oxides ($NO_x = NO + NO_2$), and sunlight, on the intrusion of O_3-rich air from the stratosphere, and on deposition onto the surface of vegetation and ground (e.g. Jenkin and Clemitshaw 2000). O_3 is *reactive*: It is destroyed upon contact with surfaces as well as in gas-phase reactions. Some of the products of these reactions have very low vapour pressures, and through them O_3 indirectly takes part in the formation of aerosols.

Aerosol particles affect the climate directly by reflecting or absorbing solar radiation and indirectly by acting as cloud condensation nuclei. Either way, aerosols reduce the amount of solar radiation reaching the surface and, consequently, 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). A typical, highly hygroscopic ambient aerosol is NaCl (Hämeri *et al.* 2001).

In addition to chemical reactions, physical, meteorological and biological factors enter into the life cycles of *multireactive* aerosol particles.

Their formation and growth are related to the properties and transport of air masses, as well as to biological activity, depending in a highly non-linear fashion on the concentrations of the nucleating and condensing vapours, temperature and relative humidity (Raes *et al.* 2000, Kulmala *et al.* 2000a, Kulmala *et al.* 2000b, Kulmala *et al.* 2001, Mäkelä *et al.* 2002, O'Dowd *et al.* 2002). Terpenes that are emitted by the vegetation are likely to lead to aerosol formation. Their own volatility is too high for direct condensation on the surface of existing aerosol particles, but through reactions with atmospheric oxidants O_3, OH, and NO_3 (Atkinson 2000), they form products with low volatility and high molecular weight 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 α- and β-pinene, is probably very involatile and is likely to contribute to secondary aerosol by nucleation or condensation (Raes *et al.* 2000, Jenkin and Clemitshaw 2000, Janson *et al.* 2001). α-pinene is a major constituent in monoterpene fluxes in Hyytiälä (Rinne *et al.* 2000, Janson *et al.* 2001).

Field measurements of particle, trace gas and other surface fluxes are rarely made continuously and simultaneously over several years. However, the annual variation of most surface fluxes in boreal field conditions is extreme, and little information on the annual behaviour and budgets can be gained from short-term measurements. Long-term field measurements encompassing periods of extreme conditions such as drought or very cold winters are essential also for purposes of global change modelling (Grelle *et al.* 1999). The aim of this paper is to present field measurements of surface fluxes (F_m, F_H, F_w, F_C, F_{O3}, F_p) made by the eddy covariance technique above a Scots pine forest in southern Finland in 1996–2001. The measurements were continuous over the six years except for F_p and F_{O3}: The time series of F_p was sporadic and the measurements of F_{O3} began in August 2001. Previously, Markkanen *et al.* (2001) have reported F_C, F_H and F_{LE} for 1996–1999. Similarly, Buzorius *et al.* (1998 and 2000) reported short-term aerosol particle data for summer 1996 and spring 1997. In this paper, we first analyse the annual and diurnal

patterns of the fluxes, present the annual cumulative NEE calculated by two different methods, and investigate the significance of local and remote anthropogenic pollution. Secondly, we focus on the comparison of fluxes and deposition velocities of CO_2, O_3, 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 multi-reactive particles taking part in chemical, physical and biological processes. Finally, we show the connection between new-particle formation events and particle deposition velocity ($v_{d,p}$).

Material and methods

Site description

The SMEAR II station is located in a homogeneous Scots pine stand (*Pinus sylvestris* L.), planted in 1962 next to the Hyytiälä forest station in southern Finland (61°51′N, 24°17′E, 181 m above sea level). As 29% of the forests in southern Finland, the Scots pine forest in Hyytiälä is of medium site quality (Vaccinium type according to the Cajander site class system (Cajander 1909) and has a typical growth rate of 8 m^3 ha^{-1} yr^{-1}. The forest is half way through the rotation time for this site type, which is about 80 years. In 56% of the forest area in southern Finland, the dominating species is Scots pine. Regeneration and growth of the forest have been performed along standard silvicultural guidelines (Peltola 2001).

The height of the dominant stand is around 14 m and the all-sided needle area 8 m^2 m^{-2} (2002). The tree biomass is 68 t ha^{-1} (above- and below-ground) (Ilvesniemi and Liu 2001). 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.

Measurements

At the SMEAR II station, the eddy covariance (EC) measurements of F_m, F_H, F_w, F_C, and F_p are

carried out in a tower at the height of 23.3 m and partly at the height of 46.0 m. F_{O3} measurements have been carried out at the height of 22.0 m in a second measurement tower since August 2001.

We calculated the EC fluxes as 30-minute averages of the covariance of vertical wind speed and the considered scalar, such as a gas concentration. The measurement system in the tower consists of an ultrasonic 3-D fast-response anemometer (Solent 1012R2, Gill Ltd., Lymington, Hampshire, England) and a fast-response infrared absorption gas analyser (LI-6262 CO_2/H_2O analyser, Li-Cor Inc., Lincoln, NE, USA). We estimated the CO_2 storage term (the accumulation of CO_2) by means of gas gradient measurements at the heights of 4.2, 8.4, 16.8, 23.3, 33.6, and 46.0 m with an infrared absorption analyser (URAS 4, Hartmann & Braun, Frankfurt am Main, Germany). Air temperature fluctuations are calculated from the speed of sound measured with the anemometer. The particle EC system consists of the above-mentioned anemometer at the height of 23.3 m and a condensational particle counter (CPC) TSI model 3010 (lower detection limit ~14 nm). A 4.5-m-long stainless steel tube (inner diameter 3.6 mm) feeds the air from near the anemometer into the CPC, which is located in a box attached to the tower. Vesala *et al.* (1998) describe the measuring system and the site in more detail, and the post-processing procedure of F_p is presented in Buzorius *et al.* (1998) and that of F_C and F_w in Rannik (1998).

The measurement system for F_{O3} in the second tower consists of a fast-response acoustic anemometer (Solent HS1199 Research ultrasonic anemometer, Gill Ltd., Lymington, Hampshire, England) and a fast response chemi-luminescence O_3 analyser (LOZ-3 Ozone analyser, Unisearch Associates Inc., Concord, Ontario, Canada). The set-up is, in principal, the same as is described for the CO_2 and H_2O system in Vesala *et al.* (1998) and so we describe only the main differences here. The anemometer is installed at the height of 22.0 m by means of the 18-m-tall tower and a 6-m-long rectangular installation mast. The box sheltering the gas analyser is installed at a height of 15 m. The (main) sample line is a 10-m-long Teflon® PTFE tube (inner diameter 8 mm). The flow rate in this line is 233 cm^3 s^{-1} (T = 273 K, p = 1013 hPa) and it

is controlled by a critical orifice. The analyser is connected to the main line with a 0.5-m-long Teflon® FEP tube (inner diameter 1.58 mm). The flow rate in this line is 16.7 cm^3 s^{-1} (T = 273 K, p = 1013 hPa), and it is controlled by a capillary inside the O_3 analyser. Keronen *et al.* (2003) has presented the O_3 measurements in more detail.

The aerosol size distribution measurements in Hyytiälä are performed with a differential mobility particle sizer (DMPS), which has a detection range of 3–600 nanometers in particle diameter. On approximately 50 days per year, the size distributions show a clear increase in the concentration of ultrafine particles (3–20 nm). Subsequently, the newly formed particles often grow to larger sizes by condensation.

From the DMPS size distribution plots, we divided the particle formation events visually into three different classes according to their strength and clarity. Class 1 events include days with an appearance and growth of a distinct nucleation mode which persists for several hours. We classify events with few new particles, a large background concentration, or non-continuous growth as class 2 events. If new-particle formation is observed but the basic charateristics such as a distinct nucleation mode or its growth are unclear, we classify the day as a class 3 event.

We calculated the annual NEE balances for 1997–2001 by first discarding unacceptable data points, then filling the resulting gaps (g) in the time series with regressions based on the remaining accepted periods, and finally integrating all F_C values over each year. We selected data with two different methods: (1) by excluding the periods with low mixing according to the criteria described in Markkanen *et al.* (2001), usually $u* < 0.25$ m s^{-1}; (hereon referred to as the "$u*$ method"; resulting amount of gaps $g_M = 16\%$ for 1997–2001), and (2) by considering only the periods with simultaneous storage flux measurements, regardless of the level of mixing, as in one of the tests by Aubinet *et al.* 2002; (the "storage method"; resulting amount of gaps $g_A = 3\%$)). We excluded also polluted wind directions ($g_{wd} = 5\%$), periods of instrument maintenance or malfunction ($g_i = 6\%$) and, because of a false, negative offset in the measurements, all data for the three winters 1997–1998, 1998–1999, and
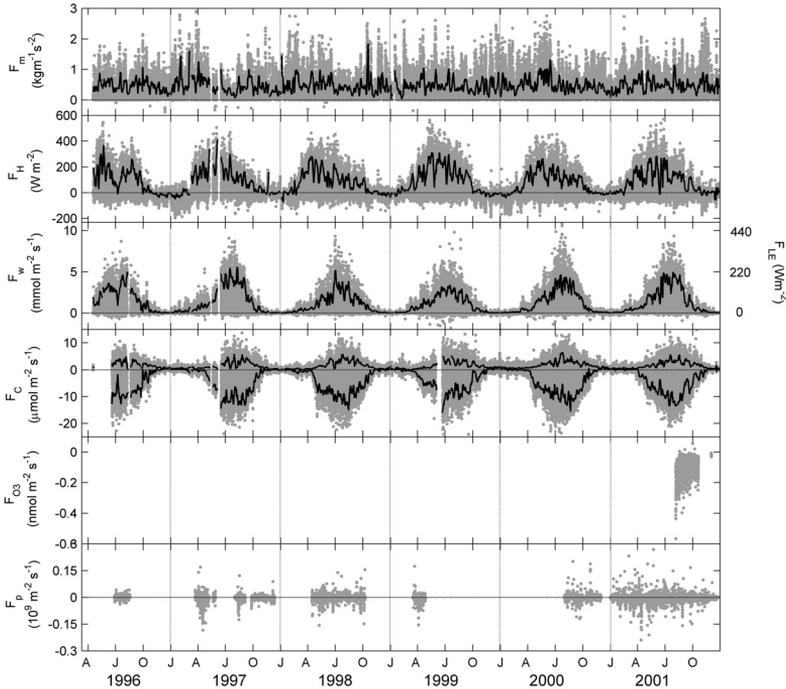
Fig. 1. Time series of surface fluxes in Hyytiälä, 1996–2001. Grey data points are half-hour averages. Black line is the average maximum flux (for 11:00–13:00, for F_C also for 01:00–03:00: the upper one) of the previous 5 days. The following fluxes were measured at 46.0 m in May 1999–June 2000 and at 23.3 m at other times: F_m = momentum flux, F_H = sensible heat flux, F_w = water vapour flux (also presented as F_{LE} = latent heat flux), F_C = CO_2 flux including storage, F_{O3} = O_3 flux (measured at 22 m), and F_p = flux of aerosol particles in the size range of 14–600 nm (only at 23.3 m). Sign convention: < 0 downwards, > 0 upwards. However, for F_m > 0 downwards.

1999–2000 (g_w = 27%). The total amount of gaps for the period 1997–2001 in the u^* method was 54% ($g_{wd} + g_i + g_w + g_M$) and in the storage method 41% ($g_{wd} + g_i + g_w + g_A$). We calculated the regressions used for filling the gaps for each spring, summer, and autumn, and for nights and days separately. The single winter-time regression was based on the winters 1996–1997, 2000–2001 and 2001–2002.

Results

According to the standard micrometeorological sign convention, we denoted downward flux from the atmosphere towards the forest negative and upward flux from the forest to the atmosphere positive. The only exception was F_m that is physically always directed downwards but is, conventionally, denoted positive. Contrary to the fluxes, the deposition velocities of O_3

and particles ($v_{d,O3}$ and $v_{d,p}$, respectively) are traditionally denoted positive when deposition occurs downwards toward the surface. Because the different sign conventions may cause confusion, throughout this paper the terms 'minimum' and 'maximum' refer to values that are closest to zero and farthest from it, respectively, regardless of the sign.

Annual patterns

Figures 1 and 2 show the entire time series of the measured surface fluxes and environmental variables in 1996–2001. In addition, for most quantities the highest daytime fluxes (11:00–13:00) averaged over the latest 5 days and denoted by a black line are superimposed on the time series. For CO_2, also the highest night-time fluxes (between 01:00–03:00, averaged over the latest 5 nights) are visible (Fig. 1).
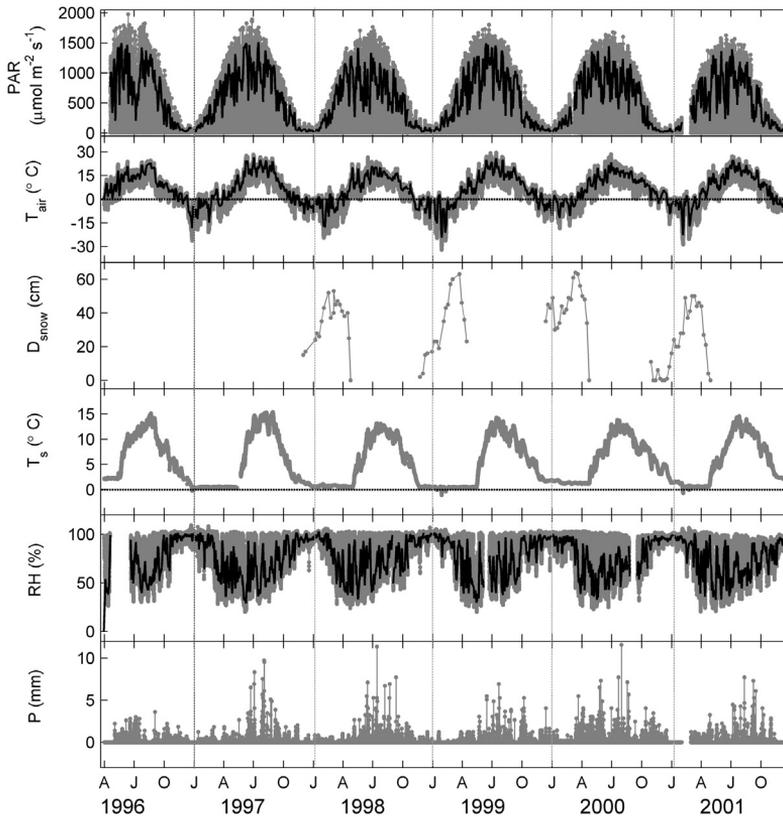
Fig. 2. Time series of environmental variables in Hyytiälä, 1996–2001. Grey data points are half-hour averages. Black line is the average maximum (11:00–13:00) of the previous 5 days. PAR = photosynthetically active radiation, T_{air} = air temperature at 8 m, D_{snow} = snow depth, T_s = soil temperature 5 cm below humus layer, RH = relative humidity at 16.8 m, P = cumulative precipitation per 30 min.

F_H, F_w, and F_C exhibited a clear annual pattern but F_m and F_p did not (Fig. 1). However, F_m was often slightly lower in July–September than at other times and generally ranged between 0 and 1.5 kg m^{-1} s^{-2}. During the summers 1997 and 1998, it was particularly low, remaining below 1 kg m^{-1} s^{-2} (corresponding approximately to u^* < 1 m s^{-1}) for 1.5 and 3 months, respectively.

F_H ranged typically from –100 to +50 W m^{-2} in winter and from –100 to +500 W m^{-2} in summer (Fig. 1). Particularly low values (–180 to –200 W m^{-2}) were observed during the winters 1996–1997

Table 1. Cumulative NEE (g(C) m^{-2} yr^{-1}) calculated by two different methods, the u^* method and the storage method.

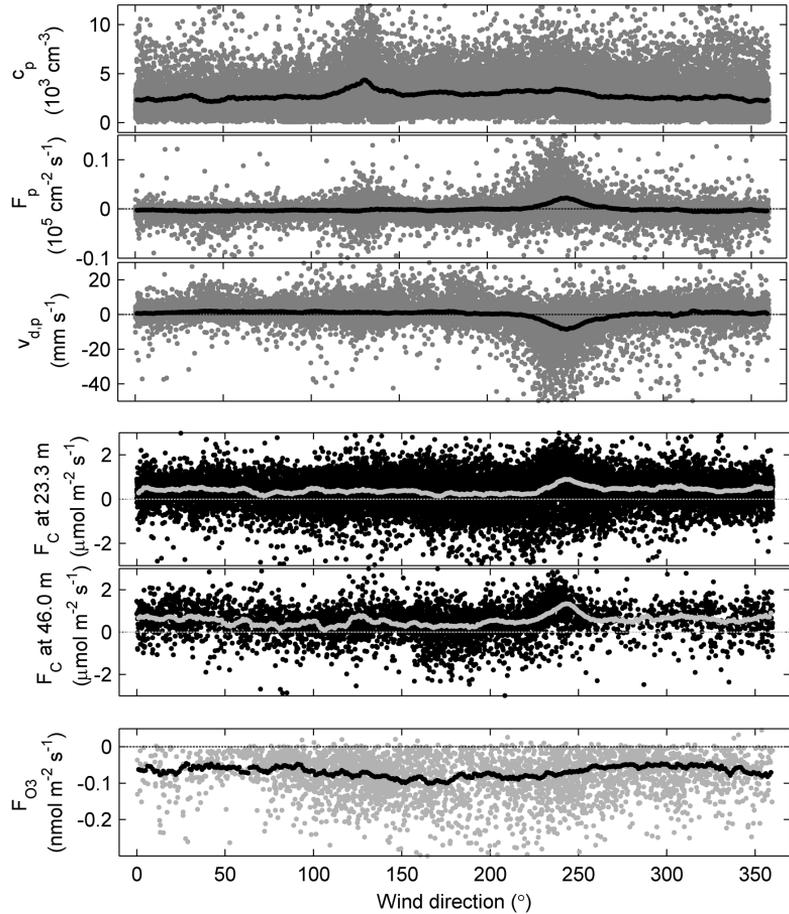| Year	u^* method	Storage method
1997	–239	–307
1998	–233	–290
1999	–162	–232
2000	–168	–214
2001	–171	–230

and 1999–2000. F_H increased very quickly in March (in 2001 already in February) and reached a maximum clearly before any other flux, already in April/May. A slow decrease started often already in July. The winter level was reached quite accurately when September turned to October, one to four weeks before the evapo-transpiration and the CO_2 uptake effectively ended.

F_w was mainly positive throughout the year, but close to zero during November–February (Fig. 1). With the first sign of snow melt F_w started to increase (Fig. 2), usually in February/March, four to eight weeks before significant CO_2 uptake began and approximately at the same time as F_H. The spring increase of F_w was more step-like and more gradual than that of F_H. In July, F_w reached maximum values of 7 to 10 mmol m^{-2} s^{-1} (corresponding to F_{LE} of 300 to 440 W m^{-2}). In autumn, it decreased quickly at the end of October or at the beginning of November.

The wintertime F_C was small and mainly positive, ranging typically from –1 to 2 μmol m^{-2} s^{-1}

Fig. 3. Dependence on wind direction of aerosol particle concentration, flux and deposition velocity (c_p, F_p, and $v_{d,p}$), of CO_2 flux (F_C) at 23.3 m and 46.0 m in winters, and of O_3 flux (F_{O3}) for August–October 2001. Points (dark grey for aerosol, black for CO_2, light grey for O_3) are half-hour averages. The superimposed curve is the average calculated with wind direction intervals of 1°.

(Fig. 1). In spring, the daytime uptake (denoted negative) started to increase abruptly, and after a time lag of a few weeks also the night-time respiration (denoted positive) increased when the snow melt was complete and the soil temperature (T_s) increased rapidly (Fig. 2). The uptake reached maximum values of –20 to –25 μmol m^{-2} s^{-1} in late June/early July and the night-time respiration maximum values of 10 to 15 μmol m^{-2} s^{-1}, again typically a few weeks later than did the uptake. The final diminishing of the daytime and night-time fluxes to the winter level happened rather simultaneously in late October–early November, at the same time as for F_w. The annual cumulative NEE obtained by the u^* method ranged from –239 to –162 g(C) m^{-2} and that obtained by the storage method from –307 to –214 g(C) m^{-2} (Table 1).

Even though no annual pattern could be deduced from a 3-month time series, it was

evident that F_{O3}, directed entirely downwards towards the forest, decreased in strength during autumn (Fig. 1). The maximum downward F_{O3} ranged from around –0.4 nmol m^{-2} s^{-1} in the beginning of August to around –0.2 nmol m^{-2} s^{-1} in mid October.

In the F_p time series the annual pattern was more difficult to discern, but both upward and downward fluxes were largest during spring (Fig. 1) when RH was at its lowest (Fig. 2). F_p ranged predominantly from –0.1 to 0.1 × 10^9 particles m^{-2} s^{-1}, that is, from –10^4 to 10^4 particles cm^{-2} s^{-1}.

Dependence on wind direction

The effect of wind direction was most evident in the particle data. The maximum particle concentration (c_p) almost doubled when the wind came
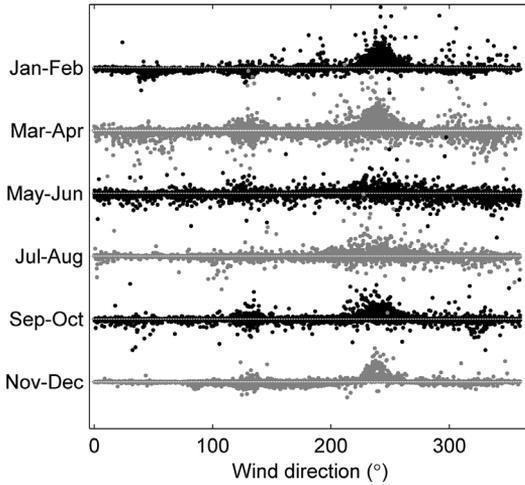
Fig. 4. Aerosol flux (F_p) *vs.* wind direction for different months, 1997–2001.

from the direction 120°–145° (Fig. 3), where the nearest settlement, Korkeakoski (distance 8 km, population of 1000, a sawmill in the town centre), is situated. Negative and positive values of F_p also increased with that wind direction. For the direction of the SMEAR II station facilities, 220°–260°, positive values of F_p increased two- to three-fold. The aerosol deposition velocity ($v_{d,p}$), defined as

$$v_{d,p} = -F_p/c_p, \qquad (1)$$

was affected only by the direction of the station facilities which produced three to four times greater negative values of $v_{d,p}$ than did the other directions. Figure 4 presents F_p for the years 1996–2001 as a function of wind direction for two months at a time. The effect of both the station facilities and of Korkeakoski almost vanished during the summer months May–August.

In F_C, a shift to the positive direction occurred in wintertime at the 23-m measurement level when the wind blew from the direction of the station facilities (Fig. 3). The effect was more pronounced at the height of 46 m: practically all fluxes were positive. We saw no effect at either height in summertime. Considering that the principal wind direction was from SW, we observed no clear wind direction dependence in the short time series of F_{O3}, either (Fig. 3).

Diurnal patterns

For comparison, Fig. 5 shows the median diurnal cycles of F_H, F_w, F_C, F_{O3}, and F_p as well as of c_p, c_{O3}, $v_{d,p}$, and $v_{d,O3}$ for August 2001. At night, F_C was quite constant at 3.5 μmol m^{-2} s^{-1}. F_C started decreasing at first daylight around 5:00 (Fig. 6). About an hour later, photosynthesis almost exceeded respiration and F_C was about to turn negative. F_C peaked at noon at –13 μmol m^{-2} s^{-1}. Subsequently, it decreased with decreasing PAR (photosynthetically active radiation, $\lambda = 400$–800 nm) and returned to the night level again at 20:30, when the last beams of light disappeared.

O$_3$ did not exhibit a plateau of constant flux at night like CO$_2$ did (Fig. 5). Instead, F_{O3} and $v_{d,O3}$ towards the forest increased already right after the midnight minimums, –0.05 nmol m^{-2} s^{-1} and 2 mm s^{-1}, respectively. $v_{d,O3}$ appeared to be at a maximum, around 7.5 mm s^{-1}, clearly before noon, between 8:00–11:00, and F_{O3} a couple of hours later, between 10:00 and 13:00, with a downward maximum of around –0.22 nmol m^{-2} s^{-1}. c_{O3} had a minimum, 18 ppb, between 6:00 and 8:00 and a maximum, 31 ppb, around 13:00–16:00.

F_p exhibited very little diurnal variation: between 12:00 and 19:00 when the relative humidity (RH) was at its lowest (50%–60%), the fluxes could be more negative than at other times, but the median was not clearly different (Figs. 5 and 6). In $v_{d,p}$, there was no diurnal variation but the general scatter was greater at daytime than at night. c_p had a weak diurnal pattern, decreasing from midnight until noon and then increasing again until midnight. Figure 7 shows some typical median diurnal cycles of aerosol particle properties for different seasons. During winter, c_p and F_p were small: In February 2001, the median c_p was about 1.5 to 3 × 10^3 (particles) cm^{-3} and the median F_p fluctuated between 0 and –1.5 × 10^3 (particles) cm^{-2} s^{-1}. However, the average level of $v_{d,p}$ was higher in winter than at any other time, around 2.5 mm s^{-1}, and the minimum and maximum $v_{d,p}$ were 1.5 and 6.5 mm s^{-1}, respectively. In May 2001, c_p was about twice as large as in February, from 3 to 4.5 × 10^3 cm^{-3} except for a short but steep minimum of 2 × 10^3 cm^{-3} between 8:00 and 12:00. In May, F_p and
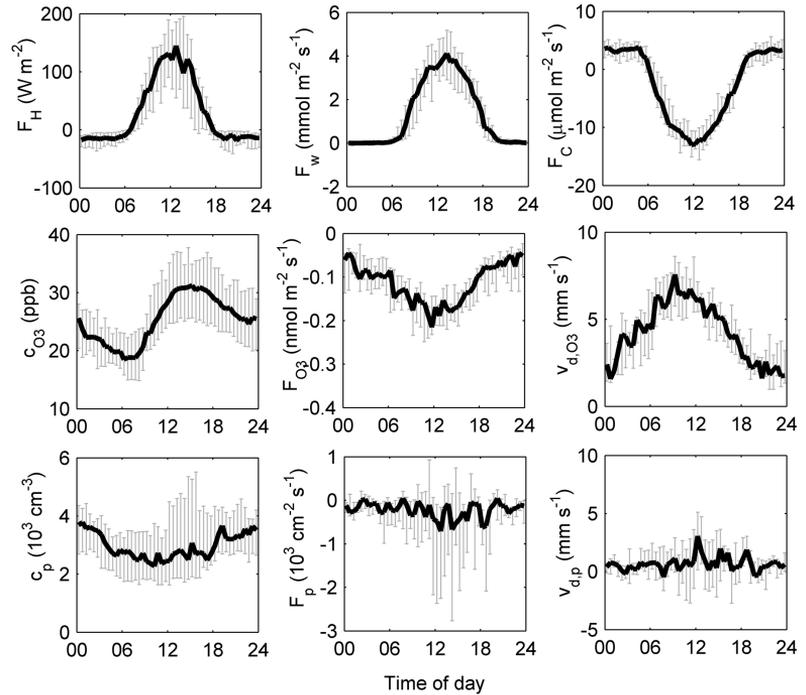
Fig. 5. Median diurnal cycle of fluxes, concentrations and deposition velocities in August 2001. Symbols as in Fig. 1. In addition: concentration of O_3 and aerosol particles (c_{O3}, c_p), deposition velocities for O_3 and aerosol particles ($v_{d,O3}$, $v_{d,p}$). Black line = monthly median of measurements for each half-hour moment, grey bars = monthly quartiles (top of the bars 75%, bottom 25%).

$v_{d,p}$ were at their largest between 12:00 and 18:00 and the diurnal pattern was very clear, although the upper and lower limits of the median F_p were the same as in February. However, the 25% and 75% quartiles in Fig. 7 show that variation of F_p around the median was considerable and that around noon, 25% of the flux was directed downwards with a rate larger than -3×10^3 cm^{-2} s^{-1}. The variation around the median was not nearly as large in $v_{d,p}$, which ranged from 0 to 5 mm s^{-1} in May. In July 1998, all three quantities were at their smallest, very close to zero. In September 1997, there was data only for a few days but the situation resembled that of May 2001, albeit on a slightly smaller scale.

Aerosol formation events

Most new-particle formation events occurred in spring under RH from 30% to 70%, many also in summer and autumn (data not shown). Figure 8 presents the connection between new-particle formation events and $v_{d,p}$. The upper pane shows that the number frequency of $v_{d,p}$ was symmetrically centred near zero on non-event days (black bars), with a mean and median $v_{d,p}$ of 0.8 and 0.7
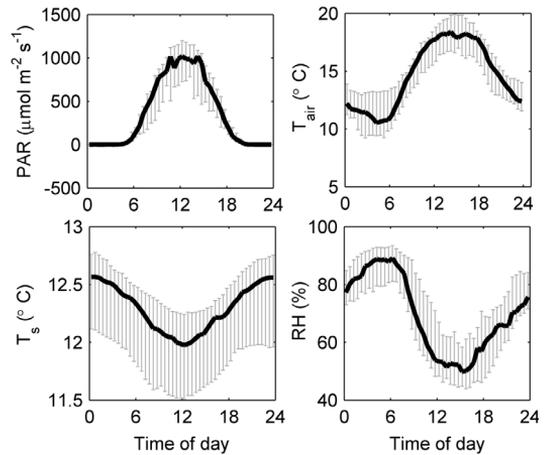
Fig. 6. Median diurnal cycles of PAR, T_{air}, T_s and RH in August 2001. Symbols as in Fig. 2. Black line = monthly median of measurements for each half-hour moment, grey bars = monthly quartiles (top of the bars 75%, bottom 25%).

mm s^{-1}, respectively. This is true also if we consider only the times when events on event days usually occur (6:15–19:45, grey bars). In this case, the mean and median $v_{d,p}$ were both 0.7 mm s^{-1}. The lower panel shows that on event days, however, $v_{d,p}$ was much more often positive than
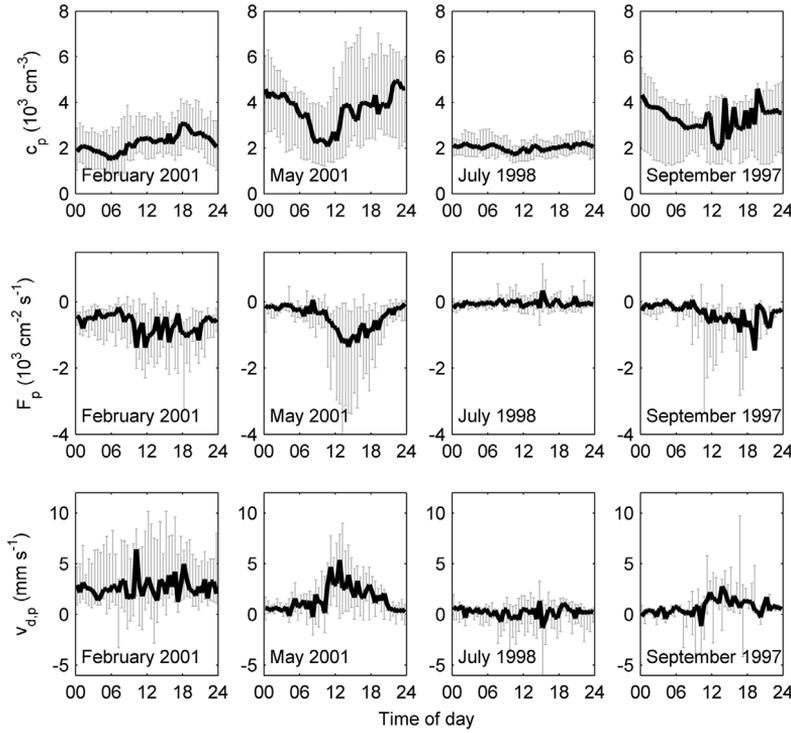
Fig. 7. Median diurnal cycles of aerosol particle concentrations, flux and deposition velocity in winter, spring, summer and autumn. Symbols as in Figs. 1 and 5.
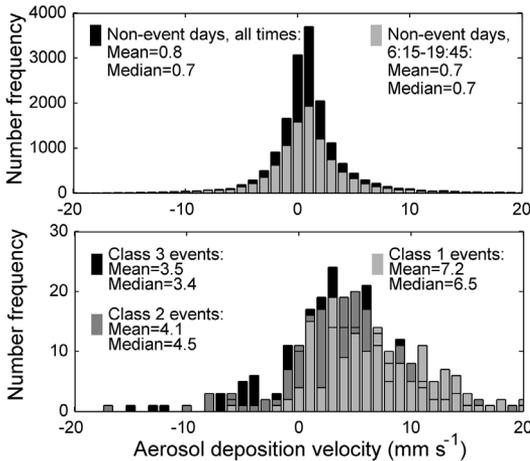

Fig. 8. Number frequency of aerosol particle deposition velocity ($v_{d,p}$) on non-event and event periods in 1996–2001. Upper pane: non-event days, all hours (black bars); non-event-days, only hours 6:15–19:45 (grey bars). Lower pane: class 1, 2, and 3 new-particle formation events (light-grey, dark-grey and black bars, respectively) (*see* Material and methods for explanation).

negative. The mean and median $v_{d,p}$ for the time periods with class 1 events were about 10 times higher than for non-event days, 7.2 and 6.5 mm

s⁻¹, respectively. The number frequency of positive $v_{d,p}$ was smaller for the less clear events, but even with class 3 events the mean and median were still 3.5 and 3.4 mm s⁻¹, four to five times as high as during non-event periods.

Discussion

Annual patterns

The variation in incoming energy and in the metabolic activity of vegetation and soil was evident in the annual patterns of F_C, F_w and F_H. The fluxes were very small in winter and markedly larger in summer. F_m did not exhibit an annual pattern probably because the wind speed and roughness of the surface do not change very much over a year in a coniferous forest. The situation could be quite different in a deciduous forest where the leaves senesce for the winter and roughness characteristics change seasonally.

The quick increase of F_H in spring is the result of increased solar radiation that heats the surface and leads to the upward transport of

warm air. When the air is still cold, the warming of the surface leads to a large temperature gradient between the forest and the atmosphere and, consequently, to a large positive F_H in spring. Until plant metabolic activity and concurrent transpiration begin, F_w is small and mainly due to evaporation from the melting snow. Consequently, the outgoing energy is primarily transported by F_H. A measure of the partitioning of outgoing energy between F_H and F_w is the Bowen ratio (F_H/F_w) that varies seasonally: In summer, increased transpiration leads to larger F_w and a smaller Bowen ratio (Markkanen *et al.* 2001).

The increase of the daytime uptake in the F_C time series in spring begins with increasing air temperatures and can vary by a month among years (Suni *et al.* 2003). A few weeks later, TER increases once the snow melts and the soil warms up, which was manifested in the time series as the increase of the night-time positive F_C. The forest was a net sink of carbon: The average annual cumulative NEE obtained by the u^* method was –195 g m^{-2} and that obtained by the storage method –254 g m^{-2}. The difference arises because the storage method assumes that advection (net horizontal transport) is negligible and does not affect the annual carbon mass balance. This is only true for terrain so flat that during low turbulence, the carbon that accumulates below the measurement level does not slide away along sloping ground undetected (nighttime problem under low turbulence conditions, for details *see* Aubinet *et al.* 2000). In the u^* method, night-time low-turbulence values are discarded and replaced with modelled values based on measurements under similar but turbulent conditions. Since advection may not be neglected in Hyytiälä because of the slight sloping of the ground, the storage method is likely to overestimate the annual uptake, whereas the u^* method probably gives the least biased estimate.

For $v_{d,O3}$, the main controlling factor during the growing season is the surface uptake resistance which, in daytime, is mainly stomatal. Towards winter, $v_{d,O3}$ decreases because the production and therefore the concentrations of O_3 are reduced and the stomatal uptake of the vegetation diminishes. Moreover, the resistances of a coniferous forest canopy and ground

to O_3 deposition are about 1.5 and 17.5 times higher with snow cover than without it, respectively (Seinfeld and Pandis 1998). Zeller (2000) reported even upward F_{O3} from the surface of snow and postulated that O_3 may be stored in the snow or in tree stands.

A large number of upward values were evident in F_p as expected from the work by Buzorius *et al.* 1998. They explained the upward flux by the existence of a particle source within or close to the canopy top. Also meteorological reasons such as concentration dilution during the early morning ABL growth (Nilsson *et al.* 2001) and roll circulation in the ABL (Buzorius *et al.* 2001) play a part. In many cases, the upward flux values result from a large random uncertainty of measurements.

Positive and negative maximum F_p were largest in spring (Fig. 1), but possible other annual patterns were effectively masked by changes in background concentration and size distribution. These changes are due to the different origins and trajectories of the air masses moving over the site, to the emissions from local and remote sources (*see* discussion on wind direction below), and to local aerosol formation.

Dependence on wind direction

The emissions from local and remote anthropogenic sources had a significant effect on aerosol particle and CO_2 concentrations and fluxes. Air coming from the nearest settlement, Korkeakoski (distance 8 km), brings with it a higher background concentration of particles. The SMEAR II station facilities are a local source in winter that increased the upward F_C at both measurement levels 1.5- to 2-fold and the upward F_p 2- to 3-fold. In summer, the effects of the station facilities and of Korkeakoski on c_p, F_p and F_C are weak because the need for heating the buildings is small and because the footprint area is smaller during the more turbulent summertime.

Especially in the case of trace gases and aerosols, the role of the more remote sources and of the air mass trajectories is important. Kulmala *et al.* (2000c) showed that the highest c_{O3} come to Hyytiälä from the clean northern trajectory sectors in winter and from the polluted, south-

ern sectors in summer. Apparently, the polluted air masses from central Europe act as a sink in winter and as a source in spring and summer when the role of photochemistry becomes more important (Kulmala *et al.* 2000c). Accumulation-mode c_p is highest in the polluted, southern air masses, and local nucleation events occurred typically in clean air masses (Kulmala *et al.* 2000a, 2000c).

Diurnal patterns

During daytime, the processes of respiration and photosynthesis act simultaneously. In Hyytiälä, optimal conditions for photosynthesis are approximately PAR > 800 μmol m^{-2} s^{-1} and 15 °C $\leq T_{air} \leq$ 20 °C. At higher temperatures, transpiration becomes too high, the stomata start to close and photosynthesis begins to decrease. Night-time TER increases with increasing T_{air} and T_s (Markkanen *et al.* 2001), but the factors affecting daytime TER are more difficult to analyse especially because we cannot infer TER from daytime NEE.

The diurnal range of $v_{d,O3}$ was similar (2–7 mm s^{-1}) in Hyytiälä as at a Scots pine forest site at Mekrijärvi in eastern Finland (0–7 mm s^{-1}, Tuovinen *et al.* 2001) and at a Norway spruce forest site in Ulborg, Denmark in June 1994 (3.5–7 mm s^{-1}, Pilegaard *et al.* 1995). In daytime, $v_{d,O3}$ is connected with PAR through stomatal uptake. That F_{O3} and $v_{d,O3}$ towards the forest increased already right after midnight when no stomatal activity was present probably reflects the reactive nature of O_3. Possible reasons for this night-time decrease of c_{O3} are surface deposition, potentially enhanced by terpenes on the surface and in the waxes of needles, and chemical reactions with NO_x or hydrocarbons such as monoterpenes. 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 concentrations increase because monoterpenes accumulate in the canopy during nights with low mixing and a shallow ABL (Rinne *et al.* 2000). The ensuing O_3 sink near the surface is sustained if the characteristic times of O_3 chemistry are fast compared with those of the low turbulent

transport. This is indeed the case according to the results of Keronen *et al.* (2003), which reveal the existence of a clear vertical profile in c_{O3} with concentrations decreasing towards the surface. Moisture, such as the formation of dew on the surface of needles and other vegetation, could also provide an additional sink inside the canopy. This hypothesis agrees well with observations (Grantz *et al.* 1995, Finkelstein *et al.* 2000, Fuentes *et al.* 1992, 1994, Lamaud *et al.* 2002, Zhang *et al.* 2002). O_3 itself is not particularly soluble and, in general, deposition on a water surface is limited by the solubility of the gas. However, if irreversible chemical reactions consume O_3 molecules inside, say, a water film on a leaf, the concentration of O_3 in the water does not increase and deposition can go on as long as the reactions take place (Sehmel 1980).

The diurnal pattern of c_p varied from non-existent in July 1998 (Fig. 7) to moderate in August 2001 (Fig. 5) and to strong in May 2001 (Fig. 7). At night, T_{air} decreases and the vapour pressures of organic hydrocarbons increase, resulting in enhanced condensation onto the surface of small nucleation-mode particles that thus grow into detectable sizes (> 14 nm). In the morning, the mixing starts and the particle concentrations are diluted in the deep daytime ABL.

A similar variation of the strength of the diurnal pattern was true also for F_p and $v_{d,p}$. During spring, F_p often remained close to zero at night and increased downwards by day as in May 2001 (Fig. 7). This kind of behaviour is typical for new-particle formation days (Buzorius *et al.* 2000). In autumn, the diurnal pattern could re-emerge for a few days but it was not nearly as clear or long lasting as in spring. Although we found no environmental variables that could explain or predict the diurnal pattern of F_p in general, in autumn its emergence seemed to be connected with the amount of radiation: In September 1997 and 2000 PAR was high and the diurnal pattern emerged, whereas in September 1998 and 2001 PAR was markedly smaller and no diurnal pattern appeared (data not shown).

The average $v_{d,p}$ was highest in winter probably because the particles are smaller in winter when the amount of condensable vapour is small (Mäkelä *et al.* 2000). $v_{d,p}$ is at its lowest for medium-sized particles with diameters between

0.1 and 1 μm because in this size range the effect of both diffusion and gravitation is small (Seinfeld and Pandis 1998). Therefore, particles tend to accumulate in this size range (accumulation mode). Outside this range, $v_{d,p}$ increases towards both smaller and larger sizes, respectively (Seinfeld and Pandis 1998, Raes *et al*. 2000).

Aerosol formation events

New-particle formation is observed around 50 times a year at the Hyytiälä site (Kulmala *et al*. 2000a). Our results show that the average $v_{d,p}$ towards the forest was greater on formation event days than on other days. This is because the nucleated particles are smaller than the more monodisperse, accumulation-mode particles that prevail during non-event conditions. This is in agreement with earlier findings based on short-term measurements in March–May 1997 (Buzorius *et al*. 2000). We found that the fraction of measurements indicating upward transport during non-event periods was 34%, during class 3 events 23%, during class 2 events 17%, and during class 1 events only 4%.

Conclusions

The results show marked seasonal variations in F_H, F_w, and F_C that stem from the seasonally varying amounts of incoming energy and metabolic activity. The effect of local and remote anthropogenic pollution is significant for c_p, F_p and wintertime F_C.

The differing properties of the fluxes were evident: The inert nature of CO_2 yields fluxes that follow environmental factors through plant physiology very closely and are relatively constant at night because, in order to be transported between the atmosphere and the biosphere, CO_2 requires the active participation of plant and microbe metabolism. Fluxes of the reactive O_3 depend on radiation in daytime, partly because of the local and regional photochemical production and destruction processes, but more importantly because O_3 diffuses into the stomata and therefore depends on the plants' environment-controlled metabolic activity. At night, O_3

concentrations, fluxes and deposition velocities change firstly because of surface deposition, enhanced by surface moisture and potentially by terpenes in the needle waxes, and secondly because of night sink reactions that are probably enhanced by the accumulation of monoterpenes near the surface. The aerosol flux, which exhibited no clear connection with any environmental variable, demonstrates the multireactive characteristics of aerosol particles that cannot be reduced into simple correlations. This resulted in practically no diurnal pattern at all, unless new-particle formation events occurred.

The formation of new nucleation-mode particles was manifested in $v_{d,p}$. The average downward $v_{d,p}$ was greater on nucleation-event days because the small nucleation-mode particles have greater deposition velocities than do the larger accumulation-mode particles that prevail on non-event days.

Further studies should include the effect of surface wetness on trace gas and aerosol deposition.

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