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TELLUS

Upward fluxes of particles over forests: when, where, why?

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ABSTRACT

Of the 60% of particle number fluxes over two forests that exceed the associated uncertainty bounds, approximately one-third are upward. These ‘apparent emission’ fluxes are not solely observed during periods when other micrometeorological fluxes are ill-defined, which implies they derive from a/multiple physical cause/s. Upward fluxes are slightly more frequent at night over the Danish beech forest but do not depend on wind direction or speed. Data from the pine forest in Finland indicate no diurnal cycle in the frequency with which upward fluxes are observed, although as in data from the beech forest the magnitude of upward fluxes is higher during the day. At the pine forest local emissions may account for some of the upward fluxes but other mechanisms appear also to play a role. Entrainment of particle depleted air from above the boundary layer, analysed via use of quadrant analysis and scalar correlations, appears to be important in the occurrence of upward fluxes at both sites. The rate of upward fluxes scales with prevailing geometric mean diameter (GMD) and consistent with the hypothesis of entrainment of relatively particle-depleted air upward fluxes appear to be associated with particle ensembles characterized by larger prevailing GMD.

1. Introduction and objectives

Particle number fluxes over forests are typically downwards (negative), but previous studies have indicated the presence of a substantial number of positive (upward) fluxes (Gallagher et al., 1997; Buzorius et al., 1998; Buzorius et al., 2000; Pryor et al., 2008b). Here, we examine the statistical significance of these upward fluxes and investigate possible explanations for this phenomenon using data drawn from two forests:

(1) The Sorø beech (*Fagus silvatica L.*) forest in Denmark. The number flux data set used in this analysis was collected between 27 May and 30 June 2004 using a TSI Incorporated condensation particle counter (CPC) 3010 sampling air from approximately 10 m above the canopy (at a height of 35 m) along with data from a Metek 3-dimensional sonic anemometer. During the field experiment, particle size distributions were measured at 31 and 43 m height using a TSI scanning mobility particle sampler (SMPS3936L25) (Pryor et al., 2007).

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(2) The Scots pine (*Pinus sylvestris L.*) forest at Hyytiälä in Finland. The particle number flux data set used in this analysis was collected between 1 January and 31 December 2004 using a TSI CPC 3010 sampling at a height of approximately 9.5 m above the canopy (23.3 m height) along with data from a Gill Solent 3-dimensional sonic. Particle size distributions were measured using two differential mobility particle sizers (DMPS) (Kulmala et al., 2001; Hari and Kulmala, 2005).

Upward particle number fluxes over forests, when observed, are typically ascribed to random uncertainty or stochastic effects in the data (e.g. Gaman et al., 2004), but at the Hyytiälä site they have also been ascribed to emissions from the nearby research station (Buzorius et al., 2000), and/or entrainment of air from above the boundary layer (Nilsson et al., 2001), and/or formation of new particles near to or within the forest canopy (Buzorius et al., 1998). Here, we evaluate the upward particle fluxes in terms of the following diagnostic analyses:

(1) We first test the postulate that upward fluxes are the result of stochastic influences and/or measurement uncertainty and hence are not physically derived. We investigate this possibility by computing uncertainties on the number fluxes using the approach of

Wyngaard (1973). We further examine whether upward fluxes are the result of high 'angles of attack' on the sonic anemometers (the angle of attack is the angle between the wind vector and the horizontal) used for the flux calculation. High 'attack-angles' are known to be more frequently observed over rougher surfaces and with lower observation heights. Hence, degradation of anemometer performance and scalar flux estimation (see van der Molen et al. (2004) and references therein) would be more likely over forests. Forest edges are generally associated with enhancement of scalar fluxes (Klaassen and Sogachev, 2006), but may also cause high angles of attack as streamlines move up over the forest edge. The Sorø research site is located in a small beech (Fagus silvatica L.) stand and is characteristic of many fragmented landscapes in Europe in that the fetch is over the forest for only 0.5–1 km dependent on wind direction, surrounding the forest the landscape is dominated by agricultural land use. The SMEAR II station is located in a relatively expansive homogeneous Scots pine stand (Pinus sylvestris L.) next to the Hyytiälä forest station, but still the fetch is homogeneous for only 250 m in most wind directions, and the location is characterized by rolling terrain. To establish if upward fluxes are associated with high angles of attack we conditionally sample the upward particle number fluxes in terms of the 'tilt' correction on the sonic anemometers.

(2) Upward particle number fluxes have been previously ascribed to particle emissions at or near the ground deriving from an anthropogenic point or line source (Buzorius et al., 2000). We investigate this possibility by conditionally sampling the particle number fluxes by wind direction.

(3) We then assess whether upward fluxes are the result of a phenomenon / multiple phenomena that exhibit a pronounced (repeated) diurnal cycle (e.g. static stability). We investigate this possibility by conditionally sampling the fluxes by hour of the day and month of the year. We further examine the fluxes in the context of prevailing wind speed, and friction velocity to determine if the upward particle number fluxes preferentially occur under conditions of weak or intermittent turbulence.

(4) The possibility that upward particle number fluxes are associated with breakdown of a nocturnal inversion was raised during the BIOFOR 3 experiment at Hyytiälä, when on several days prior to nucleation events and after a night and early morning characterized by downward particle number fluxes, a brief period of upward fluxes was observed. 'This was a frequent feature on days when the decrease in Aitken and accumulation mode number concentrations due to entrainment from the residual layer was so rapid that there were an upward flux in the air close to the surface' (Nilsson et al., 2001). We examine the possible role of breakdown of an inversion and entrainment of relatively particle-depleted air from the free troposphere using quadrant analysis (Shaw et al., 1983) and correlation coefficients between the perturbations of particle concentrations, vertical wind velocity and temperature from the sonic anemometer (Hicks, 1981). Note, we are not invoking turbophoresis (transfer as a result of gradients of turbulence intensity) as an explanation of upward particle number fluxes because particle number concentrations and fluxes as presented herein are dominated by diameters well below the range of particle sizes (relaxation times) for which turbophoresis is an effective process (Guha, 1997).

(5) Upward fluxes are related to particle dynamics occurring at or near the canopy as manifest in the prevailing geometric mean diameter (GMD). We investigate this possibility by conditionally sampling the fluxes by prevailing GMD of the particle ensemble and changes therein.

2. Methods: Flux calculation

Data from all half-hour periods when rain was observed or any instrument malfunction occurred were removed from the time series. The 10 Hz time series were then subject to despiking, detrending and co-ordinate rotation, and used to compute particle number fluxes using eddy covariance. The resulting half-hour average particle number fluxes were subject to three corrections: the Webb-Pearson-Leuning correction to correct for the variation in particle concentrations due to density variations caused by fluxes of water-vapour (Webb et al., 1980), for the attenuation of CPC response at frequencies above 1 Hz (Horst, 1997), and for the influence of correlation of fluctuations in the saturation ratio with vertical wind speed (Kowalski, 2001). It should be acknowledged that the form and applicability of these corrections is somewhat uncertain, but of these the last is typically of the largest magnitude (Pryor et al., 2008b), and it tends to reduce the frequency and magnitude of upward particle number fluxes.

Following Pryor et al. (2007), uncertainties associated with the half-hour average number fluxes derived using eddy covariance are quantified as the standard deviation on the ensemble statistics (Wyngaard, 1973) using:

$$\delta F^2 = \frac{2\Im_x}{T}\left(\overline{(w'x')^2} - \overline{w'x'}^2\right) \quad (1)$$

3. Investigating upward particle number fluxes

3.1. Diagnostics: Are the fluxes statistically robust?

Although at both of the forest sites the net particle number flux is downwards, over 40% of half-hour average particle number fluxes from Sorø and Hyytiälä are positive (upwards). Almost half of the upward fluxes (over 20% of all half-hour periods) differ from 0 by more than one-δF (Fig. 1), and approximately 10% of all half-hour periods at both sites exhibit upward fluxes in excess of two-δF. This implies that the upward fluxes are not merely a result of stochastic effects in the 10 Hz time series, and that they occur with sufficient frequency to be of interest. Further evidence that stochastic effects are not the root cause of the upward particle number fluxes may be gleaned from prior work that has shown periods with upward particle fluxes are not

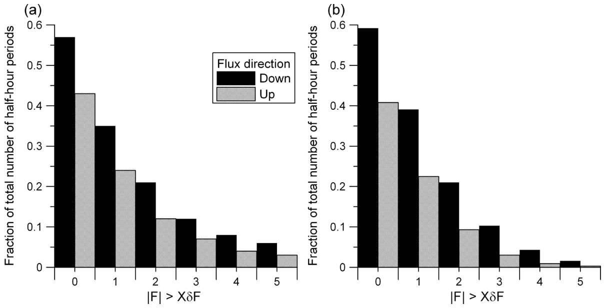

Fig. 1. Fraction of the total number of half-hour particle number flux estimates that are upward or down (*x*-axis value of 0) and where the F exceeds the specified number of multiples (X) of δF shown for (a) Sorø and (b) Hyytiälä.

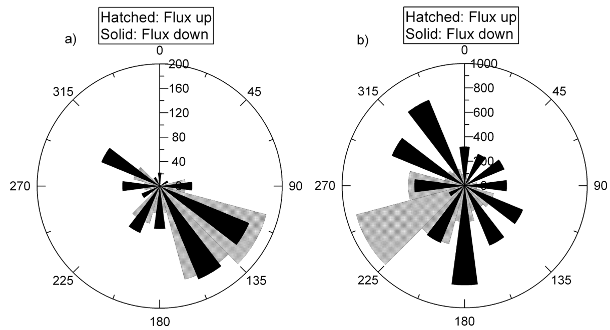

Fig. 3. The frequency with which upward and downward particle fluxes are observed conditionally sampled by wind direction from (a) Sorø and (b) Hyytiälä. Data are only included in the calculations if $|F| > \delta F$.

associated with anomalous fluxes of momentum (Pryor et al., 2008b).

High 'attack angles' on sonic anemometers are known to be associated with erroneous wind observations due to flow distortion or sheltering by the anemometer frame and hence erroneous scalar fluxes. Figure 2 shows all half-hour periods from Sorø and Hyytiälä where $|F| > \delta F$ (and in the case of Hyytiälä the wind direction was not in the sector: 220–260°, see below) in terms of the observed angle of attack on the sonic anemometer. As shown, the tilt correction at Sorø computed to correct for the angle of attack was never above 20° which is recommended as a threshold by some sonic manufacturers (Nakai et al., 2006), and only very seldom exceeded that value in data from Hyytiälä. This analysis thus implies upward particle number fluxes are not preferentially seen at high attack angles, and thus are not the result of inaccurate determination of the wind velocities and scalar fluxes.

These diagnostics coupled with the close accord between particle number fluxes computed for these sites using eddy covariance, relaxed eddy accumulation and spectral methods (Pryor et al., 2007, 2008b) thus leads to the inference that the upward particle number fluxes have a/multiple physical cause/s. We seek to determine plausible explanations for the approximately 20% of all measurement period that are characterized by upward particle number fluxes with a magnitude that exceeds the associated uncertainty (δF) below.

3.2. *Exploratory analyses of conditions associated with upward fluxes*

3.2.1. Variation with wind direction and speed. When particle number fluxes at Sorø are conditionally sampled by the prevailing wind direction in 30° sectors, downward fluxes are more frequently observed than upwards fluxes in all wind direction sectors at except in the case of the east-southeast sector in which there are slightly more upward flux cases (Fig. 3). However, the relative uniformity of the fraction of upward fluxes with wind direction implies these fluxes are not solely the product of a local ground-based particle emission source.

Data from Hyytiälä indicate a much stronger directional bias and as in prior assessments of data from this site there is a greater prevalence of upward fluxes with a wind direction that indicates potential contamination from the field station (i.e. the west-southwest direction) (Fig. 3). If the wind direction sector between 220 and 260° (based on analyses presented in Buzorius et al. (2000) as representing the directions which may be contaminated by emissions from the field station) is excluded from consideration over 1400 half-hour periods during the year (i.e. nearly 10% of all valid flux periods) exhibit upwards fluxes

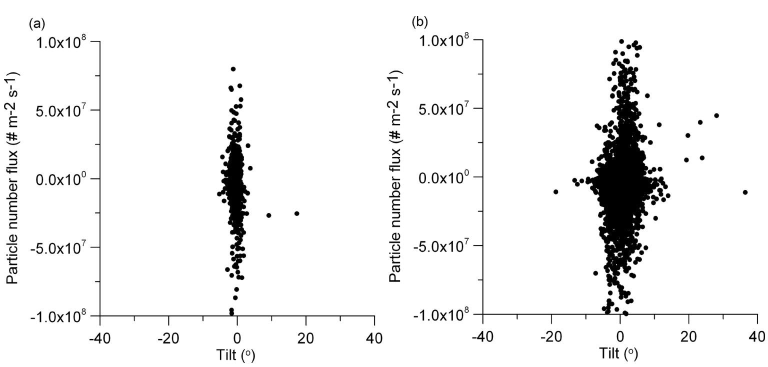

Fig. 2. Particle number fluxes (where $|F| > \delta F$ and the wind direction at Hyytiälä is not between 220 and 260°) plotted against the 'tilt' correction on the sonic anemometers based on data from (a) Sorø and (b) Hyytiälä.

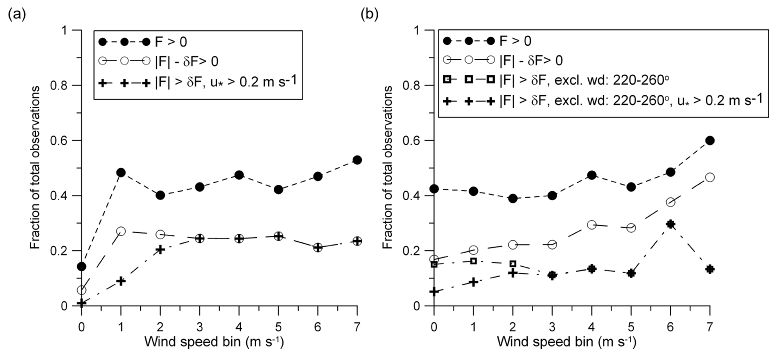

Fig. 4. The relative frequency with which upward fluxes (and upward fluxes that exceed the uncertainty) were observed at (a) Sorø and (b) Hyytiälä as a function of the prevailing wind speed. Also shown in the frequency with which $F > 0$ and $F > \delta F$ at Hyytiälä if wind directions of 220–260° are excluded from the analysis, and if an additional condition of $u_* > 0.2$ m s^{-1} is applied. Note data are only presented for wind speed classes that exhibit more than 30 members in the case of panel (a) and 100 members in frame (b).

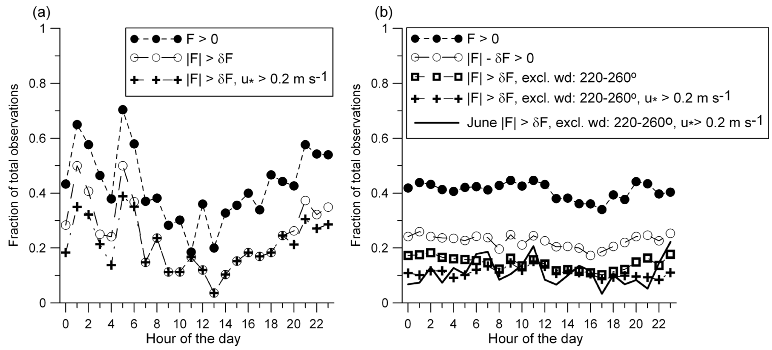

Fig. 5. The relative frequency with which upward fluxes (and upward fluxes that exceed the uncertainty) were observed at (a) Sorø and (b) Hyytiälä by hour of the day.

of a magnitude that exceeds δF. As at Sorø these upward fluxes are observed in all wind directions, and thus the apparent emission fluxes cannot be solely ascribed to local anthropogenic particle emissions deriving from a point or line source.

At Sorø there is little or no evidence of a wind speed dependence on the frequency with which upward fluxes are observed. At Hyytiälä there appears to be a tendency for upward fluxes to be observed during periods with relatively high wind speeds, but this tendency is largely absent when only cases where $F > \delta F$ are considered and the wind direction sector between 220 and 260° is excluded from the analysis (Fig. 4).

3.2.2. Diurnal/seasonal variability. Upward fluxes are observed in all hours of the day (Fig. 5), and though they are more numerous during the night at Sorø, they are of larger magnitude in the daytime. Data from Hyytiälä exhibit no diurnal bias in the frequency with which upward fluxes are observed (either over the year as a whole, or during the month of overlap with the Sorø data set [June]), though as at Sorø, the upward fluxes are typically of larger magnitude during the day.

Low values of friction velocity (i.e. $u_* < 0.2$ m s^{-1}) represent weak turbulence and have been discounted in prior research (Buzorius et al., 2001). Exclusion of these conditions leads to a decline in the frequency with which upward fluxes are observed over-night (Fig. 5) and at low wind speeds (Fig. 4).

Only data from Hyytiälä were available to examine the frequency of upward fluxes on a seasonal basis, but the results indicate maximum frequency of upward fluxes during early spring and autumn (Fig. 6). As shown, this seasonality is not merely a function of the prevalence of winds from the forest field station, or higher wind speeds in these seasons. However, at this juncture it may be noteworthy that a springtime maximum is also evident

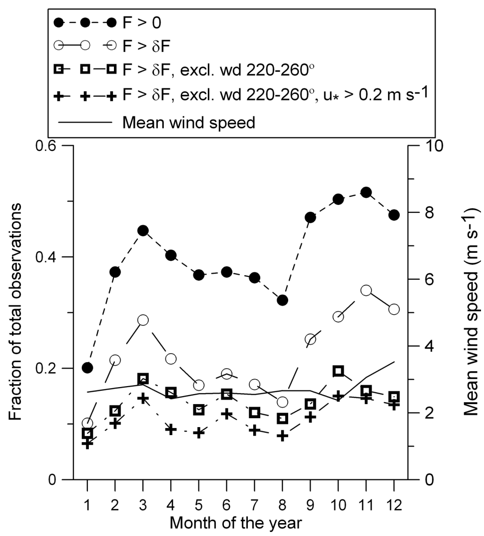

Fig. 6. The relative frequency with which upward fluxes (and upward fluxes that exceed the uncertainty) were observed at Hyytiälä during 2004 by month of the year. Also shown in the frequency with which $F > 0$ and $F > \delta F$ if wind directions of 220–260° are excluded from the analysis. The solid line depicts the mean monthly wind speeds.

in analyses of the frequency with which nucleation events are observed at this site (Dal Maso et al., 2005; Sogacheva et al., 2005).

In concluding this section it is important to recall that the data set from Sorø comprises only 1 month, while the data from Hyytiälä were collected over a year. Thus, comparisons of data from the two sites maybe confounded by seasonal variations such as phenological factors, and there is considerably larger statistical uncertainty in the conditionally sampled data from Sorø.

3.3. *Quantifying the role of entrainment*

Quadrant analysis has been previously used to examine momentum or scalar transport with respect to four different classes of transport (Shaw et al., 1983; Gao et al., 1989) based on four quadrants in the C′, w′-plane:

(1) Quadrant 1: $C' > 0$, $w' > 0$, F is positive (upward) and is the result of updrafts of particle enriched air. In the literature on momentum fluxes this quadrant is characterized as 'ejections' or 'bursts'.

(2) Quadrant 2: $C' < 0$, $w' > 0$, F is negative (downward) and is the result of updrafts of particle depleted air. In the literature on momentum fluxes this quadrant is characterized as 'outward interactions'.

(3) Quadrant 3: $C' < 0$, $w' < 0$, F is positive (upward) and is the result of downdrafts of particle depleted air. In the literature on momentum fluxes this quadrant is characterized as 'sweeps' or 'gusts'.

(4) Quadrant 4: $C' > 0$, $w' < 0$, F is negative (downward) and is the result of downdrafts of particle enriched air. In the literature on momentum fluxes this quadrant is characterized as 'inward interactions'.

Quadrant analysis was thus applied to examine the dominant processes responsible for the upward and downward particle number fluxes. Figure 7 shows one example of this analysis applied to data from Sorø and Hyytiälä for half-hour periods during which $|F| > 3\delta F$. As anticipated there is a difference in the dominant quadrants between upward and downward particle flux periods, particularly when non-zero values of the hyperbolic hole (Shaw et al., 1983) are applied. During half-hour periods characterized by downward fluxes, the 10 Hz data exhibit a tendency to cluster in quadrants 2 and 4, while during half-hour periods characterized by upward particle number fluxes, the 10 Hz data exhibit a tendency to cluster in quadrants 1 and 3. As also shown, particularly in the data from Hyytiälä, periods of upward fluxes tend to exhibit evidence of being derived primarily from 'sweeps' (downward motion of particle depleted air), rather than ejections of particle-enriched air (i.e. there are a larger number of points in quadrant No. 3 than quadrant No. 1). This implies that particularly at Hyytiälä there is a dominance of entrainment of particle-depleted air in generating periods of upward particle number fluxes. This inference is explored further below.

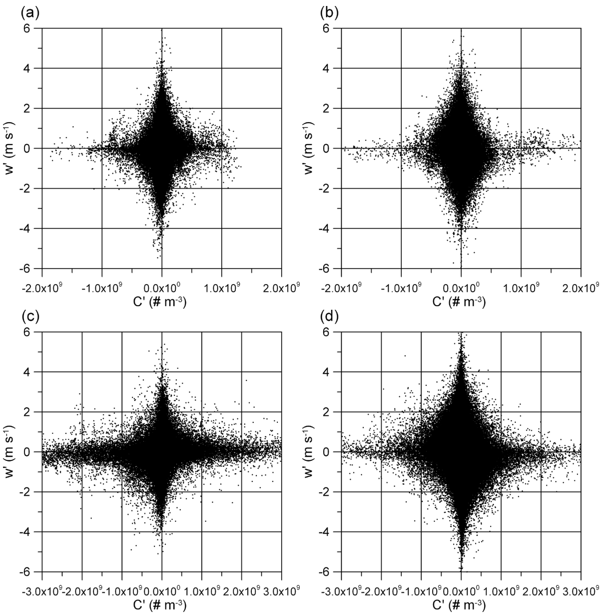

Fig. 7. Quadrant analysis of 10-Hz data (w' and C') from (a) and (b) Sorø and (c) and (d) Hyytiälä from half-hour periods when $|F| > 3\delta F$, $u_* > 0.2$ m s^{-1} and in (c) and (d) when wind directions $\neq$ 220–260°. (a) and (c) Depict data from periods of upward particle fluxes. (b) and (d) Data from periods of downward particle number fluxes. For the sake of clarity only every 9th value is plotted in (a) and (b) and only every 18th value is plotted in (c) and (d).

Observed scalar fluxes near to the ground are derived from two components; local surface-driven turbulence (also referred to a 'bottom-up' processes) and non-local or 'top-down' processes such as entrainment of air from above the mixed-layer (Sempreviva and Gryning, 2000). The importance of these components is manifest in the correlation (R_{xy}) between scalar fluctuations:

$$Rxy = \frac{\overline{x'y'}}{\sigma_x \sigma_y} \tag{2}$$

Variations in R_{xy} with different parameter combinations derive from variations (or differences) in sources and sinks of the scalars, the influence of non-local processes (these will gain importance when local production of scalar variance is small) and noise in the instrument signals.

Figure 8 shows the correlations for $C'w'$, $T_v'w'$ and $C'T_v'$ computed using data from Sorø and Hyytiälä. As expected, R_{Tvw} exhibits a bifurcation representing the variation of the surface as a source of sink of heat with the diurnal cycle. As in the Kansas data set $R_{Tvw} \approx 0.6$ in strongly unstable conditions, declining to approximately 0.35 at near-neutral stability (though the measurement uncertainty is large) and to approximately -0.35 to -0.4 in stable conditions (Hicks, 1981). Also as in prior research R_{Cw} is considerably lower than R_{Tvw}. However, due in

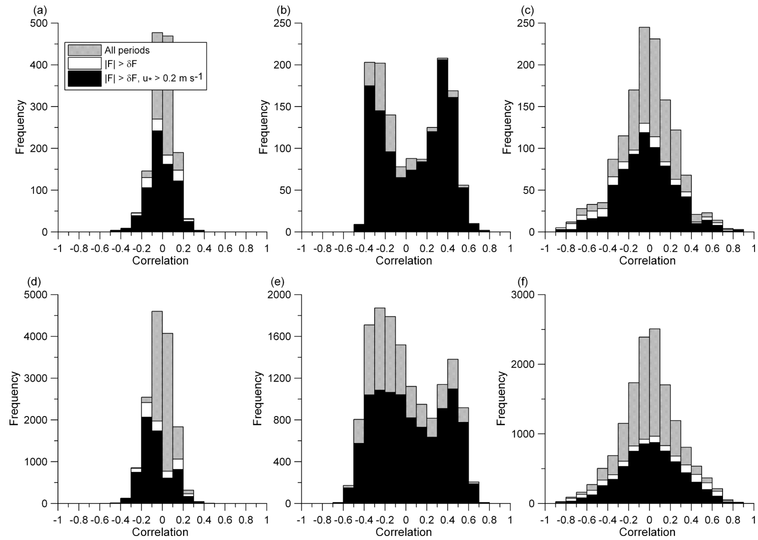

Fig. 8. Correlations (computed from the 10 Hz data over each of the half-hour sampling periods) of (a) and (d) particle number concentration fluctuations and fluctuations of vertical velocity, (b) and (e) temperature and vertical velocity perturbations and (c) and (f) particle number concentration fluctuations and temperature perturbations. Results are shown for (a–c) Sorø and (d–f) Hyytiälä. Note the *y*-axis is not consistent between frames, and that data from Hyytiälä collected during periods when the wind direction was between 220 and 260° are excluded from the analysis.

part to improvements in particle instrumentation over the last twenty years, the signal-to-noise ratio has increased and accordingly the values of R_{Cw} computed using the data sets presented herein are higher than those cited in prior research (Katen and Hubbe, 1985; Lenschow and Kristensen, 1985; Neumann and den Hartog, 1985; Duan et al., 1988), particularly when only periods of $|F| > \delta F$ are considered (Fig. 8). It should be noted that due to the large sample size (i.e. $n \approx 18\,000$, though effective n will be $\ll n$ due to the high temporal autocorrelation in the time series (Zwiers and Vonstorch, 1995)), $|R_{xy}|$ greater than $\approx$0.05 is significant at the 95% confidence level.

Half-hour average correlations between temperature and particle number perturbations, R_{CTv}, span from −1 to 1, reflecting variations in flux direction to/from the canopy, but many of the correlations are close to zero which emphasizes the complexity of relationships between transfer of heat and particles and likely to heterogeneity of the distribution of particle sources.

As discussed above, prior research (Nilsson et al., 2001) and the results of quadrant analyses suggest entrainment of particle-depleted air from above the mixed layer is a possible mechanism for causing upward particle number fluxes. In brief, the following scenario may be constructed: If relatively warm, particle-depleted air were entrained into the mixed layer (possibly as a result of the breakdown of the nocturnal inversion) this could result in; (i) top–down processes dominating scalar variance in the mixed-layer, (ii) a decoupling of temperature and particle number fluctuations since the downward motions would be associated with positive temperature excursions but negative fluctuations of particle number concentrations, potentially resulting in negative correlation coefficients (R_{CTv}) and (iii) the appearance of upward particle number fluxes. Given this type of phenomenon would likely dominate during the daylight hours, the data were conditionally sampled to include only daytime observations and periods when $|F| > \delta F$. The results from the Hyytiälä data set indicate that of the half-hour periods that fulfil the selection criteria (and are not associated with wind directions of 220–260°) over two-thirds, exhibit negative R_{CTv}. The inference may be drawn that under the aforementioned scenario, entrainment may indeed play key role in explaining upward particle number fluxes at least during the daylight hours. Similar analyses performed using the Sorø data set indicate only one-quarter of the daytime upward fluxes are associated with entrainment, possibly indicating this phenomenon is less prevalent at the Danish forest during May and June, or that Denmark experiences conditions conducive to development of elevated particle-enriched layers which may then be subject to entrainment.

Questions arising from scalar similarity are now the subject of considerable debate in the micrometeorological community and the ratio of R_{xw} to R_{Tvw} has been used as a measure of the efficiency of transport of scalar x to heat (Lamaud and Irvine, 2006). It is observed that this ratio (λ) when computed for x = water vapour concentration exhibits a profound stability dependence over forests and that generally heat is more efficiently transported than water vapour except in cases when $L \to \infty$ (i.e. in near-neutral conditions) (Lamaud and Irvine, 2006). Hence, λ was computed for R_{Cw}/R_{Tvw} for each of the forests for all cases when $F > 0$ and $F > \delta F$. As shown in Fig. 9, there is a qualitative tendency for λ to decline as $-(z-d)/L$ increases, indicating that as in the case of water vapour, heat is more efficiently transported than particles except in cases when $L \to \infty$ (i.e. in near-neutral conditions). However, although there is a tendency in λ similar to that which has been observed in R_{qw}/R_{Tvw} (Lamaud and Irvine, 2006), there is much more scatter in the relationship between λ computed using particle concentrations and heat versus stability which may indicate either (i) the greater statistical uncertainty in R_{Cw} due to the lower correlation coefficients or (ii) the influence of other confounding effects, such as the role of particle-dynamics in dictating the flux and ratio of the scalar fluctuations.

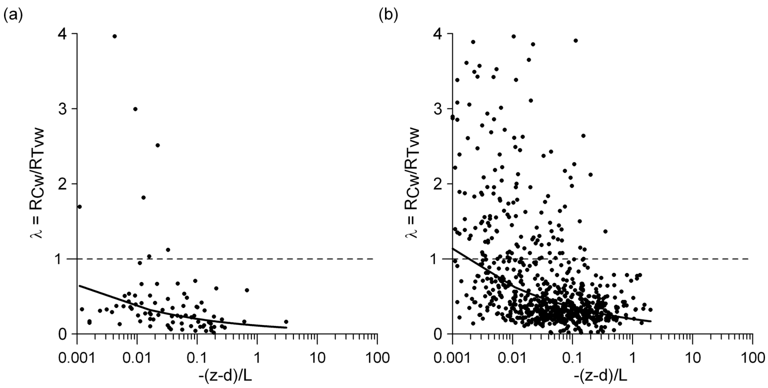

Fig. 9. Ratio of R_{Cw} to R_{Tvw} (λ) versus $-(z–d)/L$, based on data from (a) Sorø and (b) Hyytiälä. As a visual guide a power law has been fitted to the data, but this not intended to act as a quantitative description of the relationship between λ and $-(z–d)/L$. Recall that as $L \rightarrow \infty$, stability conditions are increasingly near-neutral, while for negative L, as $L \rightarrow 0$ conditions are increasingly unstable. Data are only included in the analysis if $F > 0$, $F > \delta F$, $u_* > 0.2$ m s^{-1} and at Hyytiälä if the wind direction $\neq$220–260°.

With respect to the latter it is worth recalling that processes such as coagulation and condensation have been demonstrated to be of sufficiently similar timescales to cause flux divergence (Pryor and Binkowski, 2004). The possible role of particle dynamics in generating upward particle number fluxes is further discussed below.

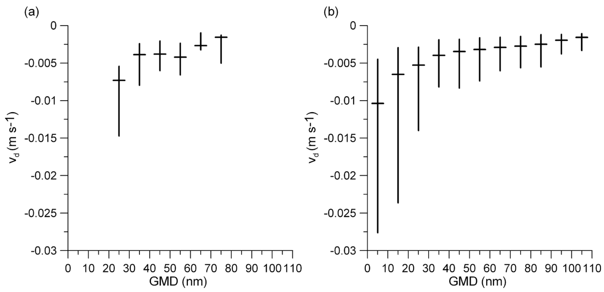

Fig. 10. The deposition velocity (v_d) during periods of upward particle number fluxes as a function of prevailing geometric mean diameter (GMD) based on data from (a) Sorø and (b) Hyytiälä. The horizontal bar shows the median value in each size bin and the vertical bars denote the inter-quartile range. Data are only included in the analysis if $F > 0$, $F > \delta F$, $u_* > 0.2$ m s^{-1} and at Hyytiälä if the wind direction $\neq$220–260°.

3.4. Evaluating links to particle dynamics

A link between particle flux directions and particle dynamics (as manifest as the prevailing number GMD or changes in GMD) can be invoked based on at least two lines of reasoning:

(1) First, the GMD represents the average particle mobility. Transfer across the quasi-laminar boundary layer above a leaf surface is demonstrably important to the total resistance to vertical particle transport. The resistance to deposition offered by this layer increases as particle diameter increases from 10 to 100 nm (Pryor et al., 2008a), and hence dictates their availability for updrafts to eject them from the canopy.

(2) Second, the GMD is a product of processes such as nucleation and growth (via condensation and coagulation, where the latter would result in a loss of particle number even in the absence of vertical fluxes). These processes may include near-canopy nucleation or growth of recently nucleated particles at/near the canopy to measurable sizes additionally changes in the particle ensemble due to processes such as nucleation will be manifest by changes in prevailing GMD.

Analysis of the particle number flux data sets with respect to the prevailing number GMD computed from the SMPS and DMPS systems, indicates that, although there is considerable scatter in the data, in accord with expectations the emission velocity (i.e. negative v_d) increases with decreasing GMD (Fig. 10). However, the deposition velocity does not appear to be linked to changes in GMD from the previous half-hour period or the prior hour (Fig. 11). Nevertheless when the data from Hyytiälä are separated into upward and downward particle number fluxes and then conditionally sampled based on prevailing GMD, there is a tendency for particle ensembles with larger GMD to be associated with upward fluxes (Fig. 12). This is consistent with (but certainly not proof of) the theory that entrainment is associated with upward fluxes. The free tropospheric air would likely contain a more aged particle ensemble with relatively low number concentrations but higher mean GMD.

4. Summary

Upward particle number fluxes over vegetated surfaces have generally been ascribed to stochastic processes and have had little or no importance attached to them. However, we present data from two forests that indicate they are statistically robust, are observed with sufficient frequency to merit consideration, and may derive from physical causes.

At the current time no definitive explanation for upward particle fluxes can be advanced. It may be that there are two or more mechanisms involved and that the relative importance of those mechanisms varies with (for example) hour of the day. Entrainment of particle-depleted air from the free troposphere as indicated by quadrant analysis and signatures in the cross-correlation of scalar fluctuations appears to be causally linked to a high proportion of upward particle number fluxes at Hyytiälä,

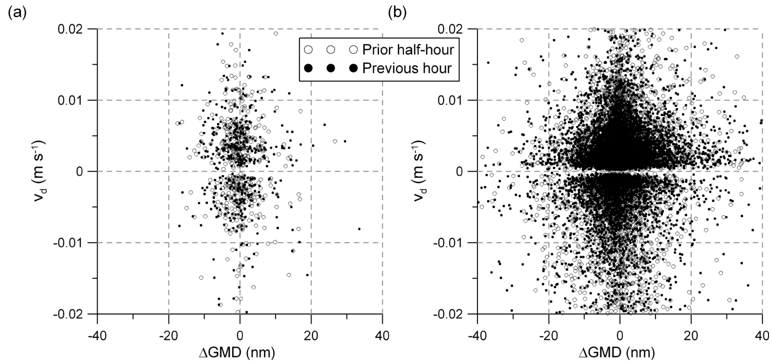

Fig. 11. Half-hour average v_d plotted against the change in GMD from the previous half-hour period, and the prior hour for (a) Sorø and (b) Hyytiälä. Data are only included in the analysis if $F > 0$, $F > \delta F$, $u_* > 0.2$ m s^{-1} and at Hyytiälä if the wind direction $\neq$ 220–260°.

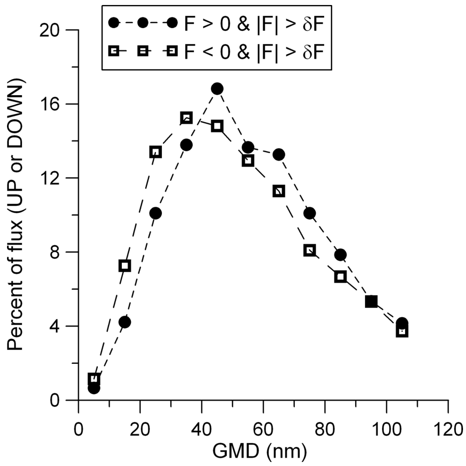

Fig. 12. Data from Hyytiälä presented in terms of the frequency with which upward or downward particle fluxes are observed according to prevailing GMD. Data are only included in the analysis if $F > 0$, $F > \delta F$, $u_* > 0.2$ m s^{-1} and at Hyytiälä if the wind direction $\neq$ 220–260°.

but explains only a relatively small fraction of upward fluxes at Sorø. While emission velocities for particle number scale with prevailing number GMD in a physically consistent manner, the occurrence of upward fluxes does not appear to depend on changes in prevailing GMD. However, periods of upward particle number fluxes at Hyytiälä appear to be characterized by higher GMD, which is consistent with entrainment of relatively particle-depleted, chemically aged air from the free troposphere.

Given the prevalence of upward particle number fluxes, further research is warranted.

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6. Appendix: Nomenclature

C is the particle number concentration
d is the displacement height
$|F|$ is the absolute value of the particle number flux (F)
L is the Monin-Obhukov length
R_{xy} is the correlation coefficient between scalar fluctuations ($x' = x - \bar{x}$, where x and $y = T_v$, C or w, $x' = x - \bar{x}$ (primed quantities indicate 'instantaneous' deviations from the half-hour mean))
T_v is the sonic measured virtual temperature
T is the integration time interval used in the eddy covariance calculations
$\bar{u}$ is the mean wind speed
u_* is the friction velocity
v_d is the deposition velocity
w is the vertical velocity
wd is the wind direction
x is the scalar of interest
z is the effective measurement height
δF is the inherent uncertainty on the particle number fluxes (F)
λ is the ratio of R_{xw} to R_{Tvw} and is thus a measure of the efficiency of transport of scalar x to heat
σ_x is the standard deviation of x
$\Im_x$ is the averaging time to determine the turbulence properties to a given accuracy ($\Im_x \approx z/\bar{u}$)

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