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Size-resolved eddy covariance fluxes of nucleation to accumulation mode aerosol particles over a coniferous forest

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ABSTRACT

In late summer 2013, size-resolved particle number concentrations above and below a coniferous forest canopy have been measured at the 'Waldstein' site in NE Bavaria (GER). The application of a fast particle spectrometer (ELPI+) on top of a 30 m scaffolding tower allowed the direct calculation of particle number fluxes for 10 separate size stages using the eddy covariance method. To the best of our knowledge this study is the first to report size-resolved and directly measured eddy fluxes and deposition velocities for a wide size range covering nucleation to accumulation mode particles with diameters between 0.006 and 1.4 μm. The results suggest that new particle formation (NPF) leads to enhanced particle number fluxes. Overall, a net deposition of −4.1e+07 particles m^{-2} s^{-1} corresponding to a mean deposition velocity of −0.27 cm s^{-1} was observed. Size dependent upward fluxes for ultrafine particles were mainly observed during the morning on NPF days. In total, about 30% of the measured fluxes were upward directed. This study examines correlations between apparent upward fluxes and explanatory variables such as random flux errors and wind direction. Further, a comparison of the measured deposition velocities to latest eddy covariance observations and models was performed. A simple second-order fit to observations for particle sizes between 0.1 and 1 μm is presented.

1. Introduction

Atmospheric aerosol particles can either deposit on a surface by mechanisms of dry or wet deposition, or they can be emitted from surfaces into the atmosphere. The resulting net exchange flux is a driver for atmospheric particle number and mass distributions. Thinking one step further, the exchange of particles has consequences for environmental impacts, namely the atmospheric radiative forcing, hazardous impacts on human health, impacts on atmospheric visibility, and nutrient and pollutant fluxes. With recent developments in the field of micrometeorology and scientific instrumentation it became popular to estimate the turbulent exchange of gases and aerosol particles with the eddy covariance technique, which is nowadays a well-established direct tool to study surface-atmosphere exchanges. In this context, natural surfaces, especially forest canopies, have commonly been described as particle sinks, whereas particle emission events have been treated as rare phenomena or as artifacts from measurement uncertainties (summarized in Pryor et al., 2008b). Based on aerosol physics, theoretical models have been proposed to determine particle deposition velocities as a function of particle size. In the last 20 years a relatively small number of eddy covariance (EC) observations of the turbulent exchange of particles over forest canopies have been conducted (reviewed in Pryor et al., 2008a). Out of these, several quantify the total particle number flux for a broad size spectrum (usually 0.01–0.7 μm) using a condensation particle counter (CPC) and derive size-resolved deposition velocities from simultaneous observations of the geometric mean diameter of the respective particle number distributions as measured by a mobility particle sizer, DMPS or SMPS (Buzorius et al., 2000; Pryor, 2006; Pryor et al., 2007; Grönholm et al., 2009). In another approach, Buzorius et al. (2001) and Held and Klemm (2006) were able to distinguish deposition velocities of ultrafine particles of a few nm of size from total deposition velocities using two condensation particle counters with different lower cut-off diameters. The limitations of this method are evaluated in Buzorius et al. (2003). Due to the lack of fast particle sizing instrumentation, Gaman et al. (2004) and Grönholm et al. (2007) applied the relaxed eddy accumulation technique in combination with DMPS measurements of particles smaller than 0.15 μm. However, direct EC-measurements of size-resolved particle fluxes over forest canopies have only been conducted three times, for particles larger than 100 nm (Gallagher et al., 1997a), for sub-100 nm

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particles (Pryor et al., 2009), and for particles larger than 246 nm (Vong et al., 2010), respectively. Based on these results parameterizations for the deposition velocity as a function of particle size and the turbulence characteristics have been proposed (e.g. Gallagher et al., 1997a; Vong et al., 2010). For a better understanding and more robust parameterizations of the particle exchange, more observations of directly measured fluxes above forest canopies – especially for ultrafine particles – are needed. With these considerations in mind, the following questions arise: do direct measurements of size-resolved particle fluxes support the current understanding of particle exchange? Do size-resolved measurements reveal apparent upward fluxes of a specific size fraction? Do upward fluxes correlate with flux errors or other meteorological variables? This study reports size-resolved fluxes for nucleation to accumulation mode particles obtained from direct eddy covariance measurements with a real-time particle spectrometer at a mid-altitude coniferous forest site.

2. Study site and instrumentation

During August–October 2013 measurements were carried out at a mid-altitude coniferous forest site (‘Waldstein’, 50.142127 N, 11.86696 E, 776 m above sea level) in the Fichtelgebirge Mountains (SE Germany), which is mainly comprised of planted Norway spruce (*Picea abies* (L.) Karst). Two independent EC-setups (reference setup and size-resolved setup) with two different particle sensors, a CPC for total fluxes, and an ELPI+ for size-resolved fluxes, were installed 7 m above the forest canopy on top of a 30 m scaffolding tower. Additional measurements of particle size distributions were carried out 2 meters above ground level at a 100 m × 100 m clear cut in a distance of about 270 m to the tower. A more detailed site description including footprint analysis is published in Klemm et al. (2006).

2.1. Total particle number fluxes, and ground-based particle number concentrations

Total particle number concentrations for particles with diameters >11 nm were sampled with a condensation particle counter (CPC 3760A, TSI Inc., Shoreview, MN, USA) mounted 31 m above ground level. Eddy covariance particle number fluxes were calculated from the CPC data and vertical wind speed measured with a Young Model 81000 sonic anemometer (R.M. Young, Traverse City, MI, USA). The inlet of the CPC sampling line had a vertical offset of 20 cm and a horizontal offset of 15 cm in respect to the measuring region of the sonic anemometer. Particle size distributions were measured using a scanning mobility particle sizer (SMPS) custom-built by TROPOS (Leipzig, Germany) according to the design recommended by Wiedensohler et al. (2012). The mobility diameter range from 10 nm to 750 nm was scanned with a time resolution of 5 min. The sheath flow was set to 5 l min^{-1} while a sample flow of 1 l min^{-1} was directed to a Model 3772 condensation particle counter (TSI Inc., Shoreview, MN, USA) for particle counting.

2.2. Size-resolved particle measurement instrumentation

Airborne particle number concentrations were measured with the real-time particle spectrometer ELPI+ (Electrical Low Pressure Impactor, Dekati Ltd., Tampere, Finland). In the ELPI+ spectrometer, sampled particles are charged by corona discharge and later separated for size using the principle of inertial classification in a 13-stage cascade low pressure (40 hPa) impactor combined with a back-up filter stage. During the collection process, the charged particles produce a current, which is proportional to the respective number concentration. For more details on the ELPI principle see Keskinen et al. (1992) and Marjamäki et al. (2000). In total, 14 individual electrometers (13 impactor stages + 1 filter stage) are sampled with a rate of 10 Hz. Ambient air was aspirated through a 1.2 m long (TYGON) intake tube of 9.5 mm (3/8 in.) inner diameter with a nominal air flow rate of 0.01 m^3 min^{-1} corresponding to tube Reynolds numbers of 1100–1300 (laminar flow; Hinds, 1999). The impactor as well as the intake tube were aligned perpendicular to the tower plane with one 160° bend at the tube inlet. Particle penetration efficiencies of the measurement setup calculated according to Hinds (1999) and Baron and Willeke (2005) were typically greater than 95%, with the exception of the filter stage (84%) and stage 14, collecting the aerodynamic diameter range from 5.8 to 8.9 μm (89%). In the case of large particles in stage 14, losses are mostly caused by inertial impaction within the bend section of the intake tube, whereas losses for the filter stage are mostly caused by diffusion. Based on these results, the raw concentrations have been corrected prior to being post-processed. The aerodynamic impactor specifications are listed in Table 1.

Finally, eddy fluxes were calculated from the ELPI+ concentrations in combination with wind measurements performed by a GR-50 (GILL Instruments Ltd., Lymington, Hampshire, UK). The ultrasonic anemometer was mounted 2 m above the tower top. The ELPI+ inlet had a vertical separation of about 0.35 m with respect to the mean measurement path of the anemometer, whereas horizontal separation was close to zero. The ELPI-EC-setup was installed in 2 m horizontal distance to the reference CPC-EC-setup.

2.2.1. Evaluation of the electrical low pressure impactor ELPI+ for the application in eddy covariance measurements

An essential requirement for the application of the direct eddy covariance (EC) method is a fast response sensor usually sampling at ≥10 Hz frequency. Prior generations of the ELPI measurement principle have been used in disjunct eddy covariance studies (Held et al., 2007; Schmidt and Klemm, 2008) with sampling frequencies of 0.2 Hz, and in one eddy covariance study (Damay et al., 2009). Apart from the sampling frequency, the electrometer noise levels as well as the counting statistics of the discrete particle detection are critical factors determining the quality of the measured signal. This is especially true when the probe is employed in environments with comparatively low particle concentrations like forests. Hence, we applied various uncertainty and noise estimation methods: (i) Uncertainties in number concentration measurements $\partial(N)$ due to discrete counting were calculated according to Hinds (1999) as

$$\delta(N) = \frac{1}{\sqrt{\Sigma}}, \tag{1}$$

where Σ is the sum of particles counted within one EC averaging interval of typically 30 min. (ii) The relative flux uncertainty $(\partial \overline{w'N'})$ due to limited counting statistics (Buzorius et al., 2003) was calculated as

$$\delta(\overline{w'N'}) = \frac{\sigma_w \bar{N}}{\sqrt{\Sigma}(\overline{w'N'})}, \tag{2}$$

where σ_w is the standard deviation of the vertical wind component [m s^{-1}], $\bar{N}$ the mean number concentration [particles m^{-3}], and $(\overline{w'N'})$ the particle number flux [particles m^{-2} s^{-1}]. (iii) We estimated the flux uncertainty due to random instrument noise $(\partial cov_{\text{noise}})$ as described in Billesbach (2011) as the ratio between the real covariance (cov_{real}) and the uncertainty covariance of w and x (cov_{unc}), where the correlation coefficient has been minimized through randomization (in this case: 24 random permutations):

$$\delta cov_{\text{noise}} = \frac{cov_{\text{real}}}{cov_{\text{unc}}} \tag{3}$$