Atmospheric Environment 190 (2018) 359–375

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Size-resolved aerosol fluxes above a temperate broadleaf forest

A. Petroff[a,c,*], J.G. Murphy[a], S.C. Thomas[b], J.A. Geddes[a,d]

[a] University of Toronto, Department of Chemistry, 80 St George Street, Toronto, Ontario, M5S 3H6, Canada
[b] University of Toronto, Faculty of Forestry, 33 Willcocks Street, Toronto, Ontario, M5S 3B3, Canada
[c] Now at Centre Régional de la Propriété Forestière d'Occitanie, La Milliasolle, BP 89, 81003, Albi Cedex, France
[d] Now at Boston University, Department of Earth and Environment, 685 Commonwealth Ave, Boston, MA, 02215, USA

ARTICLE INFO

Keywords:
Particle flux
Fine particle
Broadleaf forest
Transfer velocity
Eddy correlation
Modelling
Leaf senescence

ABSTRACT

Aerosol fluxes were measured by eddy-correlation for 8 weeks of the summer and fall of 2011 above a temperate broadleaf forest in central Ontario, Canada. These size-resolved measurements apply to particles with optical diameters between 50 and 500 nm and are the first ones reported above a temperate deciduous forest. The particle spectrometer was located on top of the flux tower in order to reduce signal dampening in the tube and thus maximize measurement efficiency. The 8-week data set extends into autumn, capturing leaf senescence and loss, offering a rare opportunity to investigate the influence of leaf area index on particle transfer. A distinct pattern of emission and deposition that depends on the particle size is highlighted: while the smallest particles (d_p < 100 nm) are preferentially emitted (55% of the time), the largest particles (d_p > 100 nm) are preferentially deposited (62% of the time). For the size bins with detection efficiency above 50% (68–292 nm), the median transfer velocity for each bin varies between +1.34 and −2.69 mm s^{-1} and is equal to −0.21 mm s^{-1} for the total particle count. The occurrence of the upward fluxes shows a marked diurnal pattern. Possible explanations for these upward fluxes are proposed. The measurements, and their comparison with an existing model, highlight some of the key drivers of the particle transfer onto a broadleaf forest: particle size, friction velocity, leaf area index and atmospheric stability.

1. Introduction

Airborne particles contribute to the radiation balance of the atmosphere and to the cycling of chemicals between the atmosphere and the terrestrial surface. In particular, the deposition of nitrogen, partly in the aerosol phase, has a fertilizing effect on plant growth in nitrogen-limited systems, but can also lead to the acidification of the soil and the eutrophication of bodies of water located downstream (Fowler et al., 2009, and references herein). An accurate evaluation of such impacts requires knowledge of the vertical transfer of particles between the atmosphere and the earth. However, the magnitude of this particle transfer and its drivers remain uncertain, despite some 30 years of modelling efforts (Davidson et al., 1982; Zhang et al., 2001; Petroff and Zhang, 2010) and improvements in particle measurements based on eddy-correlation techniques (Wesely et al., 1985; Lamaud et al., 1994; Gallagher et al., 1997; Buzorius et al., 2000; Held et al., 2007; Pryor et al., 2009; Mammarella et al., 2011; Farmer et al., 2011; Deventer et al., 2015).

The vast majority of existing particle flux campaigns by eddy-correlation over forests have been based on the determination of a total particle number flux, mostly with a Condensation Particle Counter (CPC) and the simultaneous measurement of the aerosol number size distribution through a particle sizer (Buzorius et al., 2000; Held et al., 2007; Mammarella et al., 2011). The influence of particle size on deposition can then only be inferred. Faster samplers have been developed in the last decade and measurements of direct size-resolved particle number flux are being gathered in temperate coniferous forests (Gallagher et al., 1997; Vong et al., 2010; Deventer et al., 2015) and only rarely in temperate broadleaf forest (Pryor et al., 2009, on particles smaller than 100 nm) or mixed forest (Gordon et al., 2011).

A parallel research path has been followed by the community of Aerosol Mass Spectrometry, which is studying the chemically-determined mass flux of the atmospheric aerosols (Thomas, 2007; Nemitz et al., 2008; Gordon et al., 2011; Farmer et al., 2011, 2015). Though not size-segregated, these results provide extremely valuable information on the processes involved in the atmosphere-forest aerosol exchange and composition for particles between 50 and 1000 nm. The present study is part of a scientific project studying the cycles of carbon and nitrogen in a remote temperate hardwood-dominated forest in central Ontario, Canada. Although local sources of nitrogen are scarce, regional

* Corresponding author. University of Toronto, Department of Chemistry, 80 St George Street, Toronto, Ontario, M5S 3H6, Canada.
E-mail address: alexandre_petroff@yahoo.fr (A. Petroff).

https://doi.org/10.1016/j.atmosenv.2018.07.012
Received 21 February 2018; Received in revised form 21 June 2018; Accepted 9 July 2018
Available online 07 August 2018
1352-2310/

estimates of nitrogen deposition are suspected to be among the highest in North America (Environment Canada, 2004), and observations suggest that dominant trees at this site and regionally have transitioned to phosphorus limitation (Gradowski and Thomas, 2006, 2008).

To our knowledge, this is the first size-resolved measurement campaign of accumulation mode particle fluxes over a temperate broadleaf forest. The goals of this study are to help understand the dynamics of particle exchange between the atmosphere and the forest and its main drivers. The campaign extended into the fall in order to gain insights on the influence of leaf senescence and loss on particle flux, which to our knowledge has not been documented.

2. Site and methods

2.1. Description of the study site

The site is located in Haliburton Forest and Wildlife Reserve in central Ontario (45.2866°N, 78.5387°W). Land morphology is relatively homogeneous with an undulating topography on the granitic Canadian Shield. Assessment of the turbulence homogeneity and full development was part of the flux quality control (see Section 2.6). The average annual precipitation for the area is 1050 mm and the mean annual temperature is 5°C (Environment Canada). A 32-m tall instrument tower is located at ~ 500 m above sea level on a plateau area 300 m away from wetlands and 1 km away from a lake. A 100-m steep ridge is located to the north-northwest (Fig. 1). Prior measurements at the site have included eddy-correlation and chamber-based estimates of methane flux (Wang et al., 2013), nitrogen flux (Geddes and Murphy, 2014) and eddy-correlation measurements of carbon and water vapour (Geddes et al., 2014). Flux measurements were performed for about 8 weeks from August 16th, 2011 to October 10th, 2011. A diesel generator was used to power equipment during this intensive study and was located 70 m north of the tower. The northern wind sector (± 45°) was thus excluded from the analysis.

The site vegetation consists of an uneven-aged managed forest dominated by Sugar maple (*Acer saccharum* Marsh) and American beech (*Fagus grandifolia* Ehrh.). The canopy height ranges from 20 to 25 m, with an average height of 22 m. The most recent harvest took place in 1996–1997, and the stand was affected by a large windstorm in 1995. Basal area (measured 2013) is 19.7 $m^2\,ha^{-1}$ (with bootstrapped 95% confidence limits of 17.8–21.4 $m^2\,ha^{-1}$). Leaf area index (LAI) measurements were made using litter traps positioned on a grid within 150 m of the flux tower, and seasonal changes in LAI were monitored at an approximate 2-week sampling interval using the TRAC instrument method (Chen and Cihlar, 1995). This plant canopy gap-size analyzer was walked under the canopy along transects to quantify the fraction of photosynthetically active radiation absorbed by plant canopies, the leaf area index and canopy architectural parameters. Peak 2011 LAI (two-sided) was 13, and TRAC measurements indicated a decline in LAI of 70% by the end of the measurement campaign. *Acer saccharum* and *Fagus grandifolia* made up ~95% of leaf area in the stand; other species present include *Betula alleghaniensis*, *Ostrya virginiana*, *Tsuga canadensis*, *Acer pensylvanica*, and *Picea* spp (Geddes et al., 2014).

The displacement height d is estimated as 16.5 m (Raupach, 1994; Harman and Finnigan, 2007). The roughness length is evaluated using the formula emphasized by Lumley and Panofsky (1964) under near-neutral conditions:

$$z_0 = (z_R - d)\exp\left(\frac{-\overline{u_R}}{\sigma_u}\right), \tag{1}$$

where $\overline{u_R}$ is the mean horizontal wind at the reference height z_R. The roughness length was estimated to be $z_0 = 1.5 \pm 0.7$ m.

2.2. Instrumentation

Half-hourly fluxes of momentum, sensible heat and latent heat were measured using an open path system consisting of a CSAT3 sonic anemometer (Campbell Scientific, Canada), a LI-7500 infrared H_2O gas analyzer (LI-COR), and an HMP45C temperature and humidity probe. Size-resolved particle number concentrations were measured using an Ultra High Sensitivity Aerosol Spectrometer (UHSAS, Droplet Measurement Technologies, USA, operating with a 1054 nm wavelength laser), counting particles with optical diameter between 55 and 1000 nanometers at 10 Hz. For this study, the 100 original size bins were aggregated into the 14 following bins (in nm): 055–068, 068–083, 083–102, 102–125, 125–153, 153–188, 188–231, 231–292, 292–359, 359–440, 440–541, 541–664, 664–815 and 815–1000.

The aerosol sampler was located on the top of the tower, at 32 m, in an enclosure located 4 m away from the sonic anemometer. The

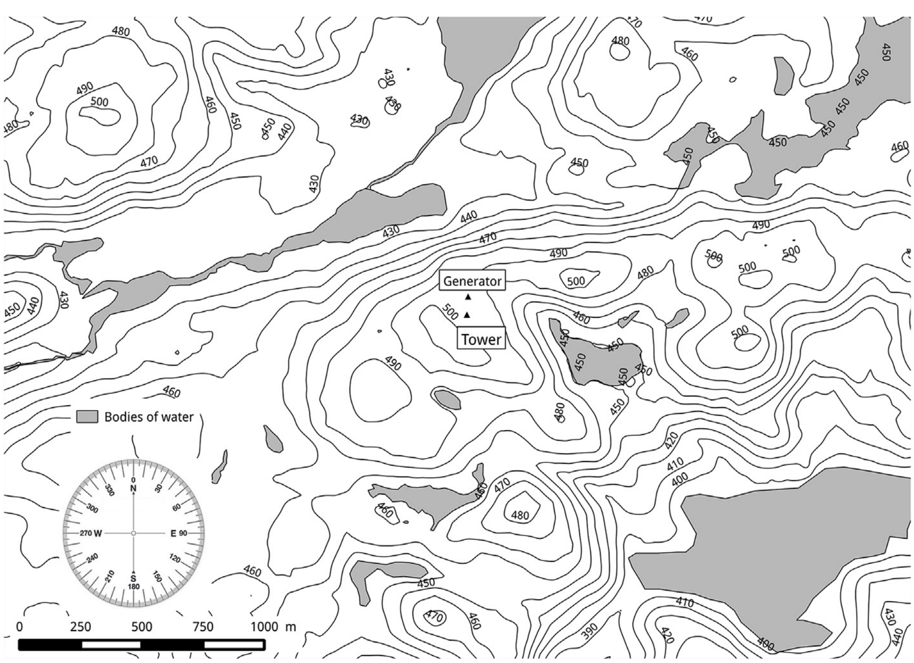

Fig. 1. Location of the flux tower. The sonic orientation is 215°. Contours correspond to altitude above mean sea level.