Appendix N:

Airborne Aerosol Flux Measurements with Eddy Correlation above the Ocean in a Coastal Environment^{*}

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Airborne aerosol flux measurements with eddy correlation above the ocean in a coastal environment

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Abstract

This work is a first pilot study of particle eddy correlation flux measurements from an aircraft using a condensation particle counter. The study reports aerosol flux measurements from an airplane conducted at approximately 33 m altitude above the ocean in the coastal region of Monterey Bay. The systematic and random uncertainties of such measurements are evaluated. Covariance was calculated for 200 s time series sampled at 10 Hz at 50 m s⁻¹ airspeed. Systematic and random uncertainties of flux measurements errors are originated from turbulence measurements errors, counter slow response time and particle counting statistics. The flux error was found to be on order of $0.1-0.3 \times 10^6$ particles m⁻² s⁻¹, for ambient aerosol concentration in the range of 1000–1500 particles cm⁻³. The direct effect of oceanic emissions (sea-salt or organic aerosol) on measured local aerosol fluxes was minimal in the studied coastal region due to ship emitted abundant anthropogenic particles.

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1. Introduction

Despite intensive research conducted during the last decade, uncertainties of aerosol impact on global climate remain high (Schwartz & Andreae, 1996). The relatively short lifetime of atmospheric aerosols and the spatial and temporal variability of aerosol sources and sinks are among the causes of this uncertainty. The need to estimate aerosol sources and sinks on local and regional scales catalyzed applications of micrometeorological methods such as the eddy correlation (EC) method. The method provides a direct estimate of vertical exchange of particles above a surface. The result is used in deriving deposition rates or estimating surface source functions. This type of measurement has been conducted in various environments: in boreal forests to analyze tropospheric aerosol (Buzorius, Rannik, Makela, Vesala, & Kulmala, 1998; Buzorius et al., 2000), above moorland vegetation (Nemitz, Gallagher, Duyzer, & Fowler, 2002), above the snow in Antarctica (Gronlund, Nilsson, Koponen, Virkkula, & Hansson, 2002), in an urban setting to correlate urban emission with traffic intensity (Dorsey et al., 2002), and in marine environment to correlate sea-spray emissions with wind speed (Nilsson et al., 2001).

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An eddy correlation measurement performed at a fixed point in space (such as a tower or an anchored ship) is limited to the footprint area somewhat adjunct to the site from the upwind direction. Measured fluxes represent vertical exchange influenced by the planetary surface within the footprint area, which could cover a distance as long as a hundred times the measurement height (Kurbanmuradov, Rannik, Sebelfeld, & Vesala, 1999). Such measurements are very useful in providing source functions or deposition rates over a square kilometer or so, but to extend the result to a larger regional scale would involve relocation of the tower, which is generally impractical or impossible. Studies on regional scales require entire networks of stations or a measurement system based on a mobile platform.

In this pilot study, we report preliminary results of particle flux measurements by the eddy correlation method, using an aircraft as the measurement platform. Advantages of the airborne platform are possibilities of making measurements in locations which are not accessible otherwise (e.g. top of boundary layer or in a power plant plume) as well as mapping of aerosol sources and sinks over a wider geographical area compared with the sampling from stationary towers. Disadvantages of this platform are related to the airplane motion: increased uncertainties in derived 3-D winds and small eddy attenuation.

The measurements were conducted in a coastal environment over the Monterey Bay from August 2003 to March 2004 during CIRPAS/NPS Twin Otter aircraft deployment in the Autonomous Ocean Sampling Network (AOSN) field campaign. Results from the entire campaign were analyzed, and a case study of the September 8, 2003 flight is presented in a more detailed manner. The average airplane speed was approximately 50 m s^{-1} while sampling at a rate of 10 Hz. There are systematic differences in the method's application if a faster airplane is used, such as increased turbulence attenuation and increased spatial averages while sampling. In this paper we focus on presenting the airborne aerosol flux measurement system. Measurement uncertainties are derived. Observed results are briefly discussed using "clean" (no ship plumes present) flux data segments. A separate paper will address aerosol fluxes in ship plumes.

2. Instrumentation and methodology

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The eddy correlation method correlates spatial averages of two time series sampled across an assembly of atmospheric eddies. In fixed-location platforms, the ergodic or "frozen turbulence" hypothesis, which states that time averages are equal to the spatial averages, is assumed to be fulfilled. The maximum length of the time series (typically 30 min for fixed-location sites) must be long enough to capture the largest atmospheric eddies as air masses are being transported downwind past the sensor, but not be so long that it is affected by diurnal or synoptic scale changes. The sampling frequency (typically 10 Hz) has to be high enough to capture the smallest eddies. The corresponding requirements for aircraft applications are discussed in Section 3. The particle flux is expressed as a covariance of the particle concentration and the vertical wind speed time series. Positive fluxes indicate upward vertical transport.

2.1. Turbulence measurements

Fast 3-D wind measurements from an aircraft require measurements of air motion with respect to the airplane and of the airplane motion with respect to the earth. The former is achieved with a radome system, which measures differential pressure drops across the semi-sphere located on the airplane nose, and the latter is achieved by inertial navigation system (INS) and global positioning system (GPS). Similar setups have been described numerous times in literature (Brown, Friehe, & Lenschow, 1983; Crawford & Dobosy, 1992; Tjernstrom & Friehe, 1991). Formulae for deriving wind speed data from the vectors of the above mentioned relative motions are discussed in Lenschow (1986) and presented in Panel 1 . However, in practice, the measured differential pressure drop deviates from the theoretical value due to the fact that the semi-sphere deviates from the ideal geometric semi-sphere, and that air in front of the airplane gets uplifted due to wing effects. Systematic differences are calibrated for by analyzing data from in-flight calibration maneuvers. The methods of calibration, corrections, and estimation of wind components with the CIRPAS/NPS Twin Otter aircraft are described in detail by Kalogiros and Wang (2002a, 2002b). Estimated random error of aircraft velocity measurements are presented in Table 1.

In-flight calibration maneuvers in four flights at different time periods during the field campaign provided data that were used to calibrate the radome system. Fig. 1 shows the results of the calibration of local radome angles from the August 15, 2003 flight. The calibration coefficients obtained were similar for all calibration maneuvers flights and were close to the ones found by Kalogiros and Wang (2002a), with the exception of the slope coefficient of the sideslip angle,

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Panel 1

$$\begin{split} U &= -U_{a} \cdot D^{-1} [\sin \psi \cos \theta + \tan \beta (\cos \psi \cos \phi + \sin \psi \sin \theta \sin \phi) \\ &+ \tan \alpha (\sin \psi \sin \theta \cos \phi - \cos \psi \sin \phi)] + U_{p} - L(\dot{\theta} \sin \theta \sin \psi - \dot{\psi} \cos \psi \cos \theta), \\ V &= -U_{a} \cdot D^{-1} [\cos \psi \cos \theta - \tan \beta (\sin \psi \cos \phi - \cos \psi \sin \theta \sin \phi) \\ &+ \tan \alpha (\cos \psi \sin \theta \cos \phi + \sin \psi \sin \phi)] + V_{p} - L(\dot{\psi} \cos \theta \sin \psi + \dot{\theta} \cos \psi \sin \theta), \\ W &= -U_{a} \cdot D^{-1} (\sin \theta - \tan \beta \cos \theta \sin \phi - \tan \alpha \cos \theta \cos \phi) + W_{p} + L\dot{\theta} \cos \theta, \\ D &= \sqrt{1 + \tan^{2} \alpha + \tan^{2} \beta}. \end{split}$$

 $U_{\rm a}$ —velocity of the air to respect to the aircraft,

 $U_{\rm p}, V_{\rm p}, W_{\rm p}$ —aircraft velocity components with respect to the earth,

 α —angle of attack, β —angle of sideslip, ψ —heading, θ —pitch, ϕ —roll,

L-distance between INS and air-sensing platform. Dot above the symbol indicates a time derivative.

Table 1

Estimated random and assumed calibration errors of input measurements in the algorithm for wind components estimation. Laboratory calibration gain error was assumed to be 0.5% for pressure and temperature measurements, and the error in gain (slope) from the calibration of local radome sideslip and attack angles using flight maneuvers was taken to be 5%

Parameter	Random error	Calibration bias error
Aircraft horizontal velocity	$0.03{\rm ms^{-1}}$	$0.1{ m ms^{-1}}$
Aircraft vertical velocity	$0.04 \mathrm{m s^{-1}}$	$0.1 \mathrm{m s^{-1}}$
Aircraft heading and roll angles	0.07°	0.1°
Aircraft pitch angle	0.05°	0.1°
Static pressure	0.07 hPa	1.0 hPa
Dynamic pressure	0.05 hPa	0.5 hPa
Sideslip and attack differential pressures	0.04 hPa	0.1 hPa
Air temperature	0.05 °C	0.5 °C

which was found to be about unity, a value higher than the previously-obtained value of about 0.9. The reason for this difference is probably an error in the past calibration of the sideslip differential pressure sensor, which in turn affected the sideslip angle calibration. Due to low noise in the sideslip differential pressure sensor (see Table 1), we were able to use the time derivative method described in Kalogiros and Wang (2002a) to compute the calibration slope for the sideslip angle, while the offset term was estimated from reverse heading maneuvers data as before.

The main sources of error in the estimation of horizontal wind components are the measurement inaccuracies in aircraft heading, sideslip angle, and dynamic pressure. Sources of errors in vertical wind components are the measurement errors of pitch, attack angle, and dynamic pressure according to the analysis of Tjernstrom and Friehe (1991). Due to the complexity of the equations (Panel 1) and iterative procedures used in the calculation of wind components, direct analytical error analysis like the differential method is difficult to apply. We used a Monte Carlo approach with Gaussian perturbation of inputs applied to measured data.

Table 1 shows the estimated average random and assumed calibration errors for the various input measurements that were used in the application of the Monte Carlo method. The random errors in GPS data are the same as the ones estimated by Kalogiros and Wang (2002a) from data when the aircraft was stationary on the ground before takeoff (static tests). Analysis showed that the total root mean square error of wind components including random and calibration gain (slope) errors was about 0.15 m s^{-1} . We note that in calculations of turbulence statistics, filtering of slow variations usually in scales above 5 to 10 km involved. In the case of vertical wind velocity, this filtering brings its average value to the expected value of about zero. After filtering, the slow-varying errors due to gain and bias errors in input parameters mentioned above do not affect turbulence statistics such as turbulent fluxes. The random error for all three wind components that remains after the filtering of large scale variations was found to be about 0.1 m s⁻¹. Reported errors have very similar values compared with earlier measurements on the same airplane (Kalogiros & Wang, 2002a) and from studies using identical turbulence measurement technique on different airplane (Tjernstrom & Friehe, 1991).

2.2. Particle counting

A modified TSI condensation particle counter (CPC) 3010 was deployed for measuring total aerosol (with sizes larger than 12 nm) number concentration. Aerosol flow rate through the counter was increased to 2.35 LPM to enhance



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Fig. 1. Calibration of (a) attack α and (b) sideslip β radome angles against free air stream estimations indicated by subscript f. The straight lines are best fit lines corresponding to the given equations.

first-order response time to approximately 0.4–0.45 s. The temperature difference between the condenser and saturator was also increased to 23 °C, modifying the D₅₀ cut-off size to about 12 nm (Buzorius, 2001). The cut-off size was verified in the laboratory prior to ambient deployment. Particle number was recorded by counting pulses during 0.1-s time intervals using NI pulse counter, model PXI 6602. The CPC was located in the airplane nose, behind the radome. The counter samples particles from the inlet, located just below the radome next to the pitot tube, through a 1-m long conductive tubing (TSI, part # 3001788), with 4 mm inner diameter. Aerosol flow in the sampling line was in the turbulent regime (\approx 7.5 lpm flow rate) to reduce signal attenuation (Buzorius et al., 2000). To compensate for the flow difference between the inlet and CPC sampling flow rates, a bypass flow parallel to the CPC was installed.

2.3. Additional measurements

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A TSI CPC 3025 was also installed in the airplane cabin. Particles to the cabin CPC were delivered using the main airplane inlet. The counter data was used for quality and consistency check of the CPC installed in the airplane nose. During August 4 to September 12, 2003, the Caltech dual scanning DMA system (Wang, Flagan, & Seinfeld, 2003) was installed on the Twin Otter. The system provided two aerosol size distributions every 73 s for the aerosol size range between 10 and 800 nm at "dry" and "humid" conditions. Relative humidities in the DMAs were maintained at < 20% and about 70%, respectively. Size distribution data could not be used in the flux calculations due to the slow response time of the instrument. They could, however, be used to bound the aerosol chemical composition by studying the aerosol hygroscopic properties imparted by the relative shift between the dry and humid size distributions.

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Other fast measurements of meteorological quantities used in this study are air and sea surface temperatures (SST) and dew point, which are measured by Heitronics KT19.85 infrared thermometer and dewpoint hygrometer, respectively. Air temperature was measured with a Rosemount platinum resistance thermometer and was corrected for the compressibility effect as part of the wind estimation algorithm. The radiometric sea surface temperature was corrected for an offset (possibly originated from the sensor dependence on altitude) of about 0.5 °C that was estimated from near sea surface data by assuming that average sensible turbulent flux should be zero for near-zero difference between sea surface and air potential temperatures. This was also confirmed after a comparison of the radiometric SST with AVHRR SST satellite data.

3. Flux calculation and uncertainties

When sampling from an airplane, the required length of scalar time series for covariance calculations is significantly reduced due to the airplane motion. It takes three minutes to transect a 9-km distance at 50 m s^{-1} airplane speed. The same air mass would be sampled by a fixed-location sensor in a 30-min time period if winds were 5 m s^{-1} , but this would provide only a single flux data point. Airborne sampling provides a quasi-spatial averaging directly. To fulfill the ergodic hypothesis, turbulence has to be "frozen" for a 3-min time period for airplane measurements as opposed to 30 minutes for tower measurements. The actual time interval for the aircraft platform (three minutes in the above example) depends on airplane altitude, speed, and atmospheric stability. During this study, time intervals of 60, 90, 120, 200 and 300 s were also considered.

For the measurement platform moving at 50 m s^{-1} speed and sampling at 10 Hz, the system can resolve eddies as small as 10 m. The fraction of eddies smaller than 10 m increases if sampling is done at decreasing altitudes (Kaimal & Finnigan, 1994). In different field deployments of the CIRPAS Twin Otter airplane, 40-Hz sampling was used for turbulence measurements. Results from those projects have shown that for runs at 33 m and higher altitude, the contribution to the latent heat fluxes from frequencies above 5-Hz (i.e., the Nyquist frequency of 10-Hz sampled data) were less than 1% (Khelif, UC Irvine, private communication). Therefore, 10 Hz sampling was adopted.

Linear trends were removed from temperature and aerosol time series for each of the time intervals prior to calculating fluxes. Vertical wind speed and the de-trended aerosol data were shifted in time to adjust for a lag caused by the particle transport from the sampling line inlet to the counter. The time lag was calculated by searching for the maximum correlation between the vertical wind speed and aerosol data. It was found to be 3.3 s.

Momentum, heat and particle fluxes were calculated using the covariance formula. Additionally, friction velocity, Monin–Obukhov length, mean wind speed, and wind direction were derived. Traditionally for fixed-location platforms, for each consecutive flux value, the time interval during which the covariance is calculated is incremented by 30 min, or the length of the time interval itself. Thus, fluxes are calculated for periods t to t + 30 min, t + 30 to t + 60, t + 60to t + 90, and so on, where t is an arbitrary starting time. For airborne applications, a different approach was adopted. Instead of incrementing by the time interval (three minutes or 200 s as described above), the increment of one second was used. Thus, fluxes were calculated for periods of t to t + 200 s, t + 1 s to t + 201 s, t + 2 s to t + 202 s, and so on. The motivation was that because of the fast airplane motion, long time steps between data points can lead to significantly different footprints (in tower measurements, footprint area depends on meteorological conditions only) for each flux data point and abrupt changes in fluxes. These changes might be difficult to interpret and relate to surface properties later on. Note that if the increment is smaller than the integral scale of turbulence, the sequential flux data points are not statistically independent.

Flux measurement uncertainties are inherited from uncertainties in turbulence sampling and aerosol number counting. Systematic error is originated from the slow CPC response time. The correction can be calculated using following formula (Horst, 1997; Buzorius et al., 2000):

$$\frac{F_{\rm m}}{F} = \frac{1}{1 + (2\pi n_{\rm m}\tau_c u/z)^{\alpha}},$$

where $n_m = 0.085$ for neutral and unstable stratification, F and F_m are total and measured fluxes, (particles m⁻² s⁻¹), τ_c is the time response constant of 0.45 s, z is the measurement altitude (m), α is 7/8 for neutral and unstable stratification, u is the true airspeed (m s⁻¹) for airborne applications and wind speed during sampling from tower. The above formula

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assumes certain shapes for concentration and vertical wind speed co-spectrum based on Kansas experiment (Kaimal & Finnigan, 1994) which is not necessarily a valid shape for measurements in other places.

Flux correction due to the slow counter response time depends on the speed at which the measurement platform is moving across air parcels. At higher speeds, larger fraction of eddies is unresolved (other in literature used terms for the same phenomenon are attenuation and smearing) for the same limited instrument time resolution. For instance, for tower sampling at 5 m s⁻¹ wind speed and time response constant 0.45 s, the flux correction is about 5% according to the Horst formula. However, during the sampling from an aircraft flying at a velocity of 50 m s⁻¹, the correction is about 30% for the same measurement altitude and the same co-spectrum shape. If the instrument time response constant is 0.05 s, corrections are 1% and 6% for tower and airborne sampling, respectively. Very fast instruments sampling at 10 Hz provide adequate flux data for airborne applications. Low-time-resolution counters lead to higher uncertainties if used to sample from fast-moving mobile platforms. The uncertainty can be reduced by sampling at the higher altitudes since with increasing the altitude, *z*, the ratio in the above formula approaches the unit.

Concentration time series measured by the CPC can be corrected for the smearing effect once the time response constant is known. It is essentially a deconvolution procedure, and there are different approaches reported on how to simplify the procedure. Kalogiros and Helmis (1993) reported a method for "speeding" time series from a slow response sensor. Others (for instance, in fast aerosol size distribution measurements by Collins, Flagan, & Seinfeld, 2002) "desmear" concentration data in order to retrieve fast variations. Collins and co-workers rewrote the deconvolution integral into a linear matrix equation.

The process of desmearing or speeding-up the particle concentration elevates the peaks and deepens the valleys in the concentration time series, making it appear more noisy. It mainly increases fluctuations at time scales of few seconds and faster. The same time running averages with time window of several tens of seconds remain unchanged. It is assumed that the additional noise introduced numerically on the desmeared concentration signal is not correlated with vertical wind speed. Both approaches gave very similar results in flux data during this study.

Random errors are associated with an error in vertical wind speed measurement (Table 1) and aerosol number counting uncertainty. For EC-derived fluxes, typically the major error source is the discrete nature of particle counting. Both random errors are estimated using differential method and values are given in the results section below. Flux uncertainties due to counting uncertainty and due to errors in measuring vertical wind speed were estimated from following formulas, respectively (Buzorius, Rannik, Nilsson, Vesala, & Kulmala, 2003):

$$\delta(\overline{wc})|_{\text{conc}} = \frac{\sigma_w c}{\sqrt{Nc Q\Delta t}},$$
$$\delta(\overline{wc})|_w = \frac{\sigma_c \delta w}{\sqrt{N}},$$

where σ_w and σ_c are vertical wind speed and aerosol number concentration standard deviation per time interval (i.e. 200 s), δw is the error in vertical wind speed measurement (0.1 m s⁻¹), Q is the aerosol sampling flow rate, (39.16 × 10⁻⁶ m³/s), Δt is the inverse of the sampling frequency (0.1 s), N is the number of samples per time interval (2000 for a 200-s time interval), c is the aerosol concentration averaged per time interval, (particles m⁻³). An additional random error in particle flux estimation is introduced due to the stochastic nature of turbulence (Rannik & Vesala, 1999).

4. Sampling site

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Each flight lasted about 4.5 h and typically started before noon Pacific Time (-8 h from GMT) depending on the weather conditions at the Monterey airport (point A in Fig. 2). After take-off, the airplane climbed over, or some days flew around, the Monterey Peninsula, then descended to an altitude of approximately 33 m (point B), and performed a series of horizontal legs in the SW-NE direction as shown in Fig. 2. After about 3 h, the airplane reached point C, flew in a triangular pattern north of it and spiraled up to approximately 1200 m elevation just offshore. From there, with a straight heading, the aircraft descended to point D (elevation 33 m), climbed up towards point E (elevation 1200 m), spiraled down to the 33-m altitude, and continued horizontal level flights towards D, F and G, respectively. At point G the airplane spiraled up to 1200-m altitude, descended to 33-m altitude flying at straight heading towards point F, climbed and descended towards point H at the shore, and returned to the base at Marina airport, point J. Data were



Fig. 2. A typical flight track during the field campaign. It is also an actual track performed on September 8, 2003. The continent is East of the thick line.

sampled continuously between points B and H. The bold line in Fig. 2 illustrates the shoreline—the right side of the line being the continent. In total there were three spiral soundings (points C, E and G) and four ascents/descents with a straight heading (C–D, D–E, G–F and F–H). If clouds were present, maximum altitudes were reduced to the cloud base elevation to avoid flying inside the clouds. Similarly, the legs were shortened in areas with heavy fog. Flight portions from points B to C and E to G were sampled while the airplane flew at an elevation of 33 m altitude.

Note that turbulence measurements are considered to be reliable only at straight heading, horizontal level flights. High roll angles occurring during the spirals and sharp turns increase errors due to uncertainties in the calibration of the radome. Typically, turbulence data from such periods are ignored. Particle fluxes in this study are thus presented only from the horizontal level flights.

5. Results and discussion

In total, 25 flights were analyzed for this study. Results demonstrated a unique spatial variability in particle vertical fluxes for each flight. Data from the flight on September 8, 2003 are analyzed in a detailed manner to demonstrate the airborne EC performance and the spatial variability of the aerosol flux. The latter is linked to the local meteorology and local aerosol sources (ship traffic). The day was chosen due to higher winds compared with the average wind speeds during the entire field campaign, available aerosol size distribution measurements, and fully functioning EC system. Standard deviation of vertical wind speed measured at 30-40 m elevation during the case study varied from 0.15 m s^{-1} close to the shore locations to 0.6 m s^{-1} at offshore locations.

5.1. Local meteorology from measurements

Fig. 3 shows wind patterns measured during the September 8, 2003 case study. The wind direction demonstrates a typical air flow in the California coastal region, which is along the coastline with wind being frequently from the northwest. The wind speeds were slightly larger compared with study averages, reaching 16 m s^{-1} .



Fig. 3. Horizontal wind speed spatial variability on September 8, 2003.



Fig. 4. Photo of the sea surface on September 8, 2003.

There were only few small drifting clouds on that day. In northwesternly flow conditions, wind speed was typically reduced inside the bay to $3-5 \text{ m s}^{-1}$, while at the mouth of the bay it was about 8 m s^{-1} . The wind also turned westwards surrounding the northern cape or northern part of bay mouth.

Fig. 4 shows a digital photograph of the sea surface taken on the same flight at the bay mouth with the camera facing down from the airplane at 30° below the horizon. Patches of whitecaps can be observed at a coverage area of about few percents. In the literature, whitecap area fraction has been found to be a function of wind speed (Monahan & O'Muircheartaigh, 1980) with typical values of about 0.5%, 1%, and 4% for wind speed of 8, 10, and 15 m s⁻¹, respectively. Additionally, sea spray depends on sea surface temperature, surface films and wave field. At open sea far offshore, Wu (1988) found whitecaps coverage to be a function of friction velocity. In coastal zones, whitecaps coverage depends on the fetch (Lafon, Piazzola, Forget, Le Calve, & Despiau, 2004). Sea spray emissions occur in areas with whitecaps. According to wave spectrum data from the National Data Buoy Center buoy off Monterey Bay, on September 8 the swell was very weak, and sea waves were generated locally by wind.

Vertical profiles of virtual potential temperature and aerosol number concentration sampled on September 8, 2003 are presented in Fig. 5. The height of the mixed marine boundary layer (MBL) gradually decreased approaching the shore (see top panels from left to right, Figs. 5a–c). At the most western point E in Fig. 2, the mixed MBL height was about 500 m. The descent from C to D indicated that the height at about 350 m (Fig. 5b), and at the shore point C 220 m (Figs. 2 and 5c). At the southern soundings (G, F and H), the reduction in MBL height was more pronounced. For

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Fig. 5. Vertical profiles of virtual potential temperature in top panels a, b and c and CPC measured aerosol number concentrations in bottom panels d, e and f. The most offshore locations are on left (a and d) panels, data from close to the coast are on the right side panels c and f and data from intermediate regions plotted in middle panels b and e.

every sounding approaching the shore, the height was found to be decreasing from 530 to 440 m, then to less than 70 m (Fig. 5c) at H as the presented profile started at the capping inversion. The mixing was mainly wind shear-driven rather than buoyancy-driven with flux Richardson number being around -0.5. The significant decrease in the mixing boundary layer height approaching the shore close to Monterey is primarily due to the wind patterns. Similar observations were reported by Winant, Dorman, Friehe, and Beardsley (1988) and Edwards, Rogerson, Winant, and Rogers (2001). They found that wind speed, wind shear and marine MBL depth change dramatically near coastal capes. In their case study, the depth of the MBL dropped from 600 to 100 m after the wind turned around the cape. During our measurement campaign, such wind turns around the cape occurred frequently under NW wind depending on the synoptic flow pattern at the northern part of Monterey Bay near Santa Cruz. This was observed as locally significant change of wind speed, negative heat flux, and low surface pressure due to the drop in the boundary layer height. By the shore in Monterey Bay, winds (Fig. 3) and turbulence (not shown) were significantly reduced.

In addition, an increased local subsidence below 800 m altitude near the shore, probably due to topographic effects from the nearby Santa Cruz mountains to the north, led to such a shallow marine boundary layer which cannot get deeper by the weak buoyancy. This was concluded by the comparison of potential temperature profile near the shore at the point H with the offshore profile. The wind direction above 500 m altitude was from the north according to the wind profile at point H (not shown).

The Monterey Area Ship Track (MAST) experiment was conducted over about the same area but further (by ~ 100 miles) offshore in June 1994, with the objective to analyze ship tracks observed in satellite images (Durkee, Noone, & Bluth, 2000). They observed MBL heights in the range 200–1000 m. Wind-driven ocean upwelling causes high spatial variability and reduction in the sea surface temperature close to the shore, sometimes reversing the atmosphere-sea surface temperature gradient and changing the stratification. At off-shore locations, mixed MBL height increased.



Fig. 6. Comparison of turbulent sensible heat fluxes H_s estimated with the eddy correlation method (averaging length 5 km) and a bulk method (COARE 3.0) on September 8, 2003.

Vertical soundings of the aerosol number concentration indicated a number of layers with elevated concentration, as shown in Figs. 5d–f. Aerosol layers aloft of the MBL were decoupled from the surface and did not affect flux measurements at approximately 33 m altitude. There are a number of layers with enhanced aerosol concentration within the MBL. Fig. 5f indicates an aerosol layer with tenfold increase in aerosol number close to the coast (spiral at C and descent to H). Aerosols were well mixed throughout entire MBL only in the most western soundings (Fig. 5d), the furthest from the coast. At intermediate distances from the coast (descent from C to D, descent to F, and ascent from F, Fig. 5e), aerosols were not homogenously mixed within the atmospheric MBL. Note that abscissa scale of Fig. 5d–e are identical.

5.2. EC performance test I: Heat flux balance

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Performance of the flux measurement system was analyzed by comparing heat flux measured by eddy correlation with the bulk heat flux derived from the difference between air temperature and radiometric surface temperature. A closure between the heat fluxes would indicate that on the overall, the radome-derived 3-D wind data represent atmospheric turbulence well and is sufficient for the eddy correlation calculations despite of the resolution limitations mentioned above. Fig. 6 shows a scatter plot comparing the sensible heat fluxes for the case study flight estimated with the eddy correlation (in the abscissa) and with the commonly used bulk algorithm COARE 3.0 (Fairall, Bradley, Hare, Grachev, & Edson, 2003), which uses average wind speed, air temperature, and sea surface temperature data (in the ordinate). The choice of the averaging length of about 5 km (100 s) was based on an ogive (cumulative sum) analysis of the co-spectra. The comparison shows a general agreement of the heat fluxes derived from the two methods for this flight with a zero bias at a 95% confidence according to *t*-test analysis (Bendat & Piersol, 1971).

A similar analysis using data from the entire field campaign was reported in Kalogiros and Wang (2004). Earlier reports in the literature comparing measurements from airborne and surface platforms also indicated an agreement between EC and bulk techniques. For example, Beardsley, Enriquez, Friehe, and Alessi (1997) compared wind and wind stress measurements from aircraft and buoy measurements off the California coast and found good agreement. Nevertheless, comparison of aircraft turbulence measurements with surface-based in situ measurements is generally difficult due to differences in sampling considerations of EC and bulk estimates (Mahrt, 1998).

The general agreement demonstrated in Fig. 6 indicates that the airborne heat flux measurements provide similar results as from bulk methods. However, this type of test is insensitive to the higher end of the frequency spectrum,



Fig. 7. Sample of FFT analysis. See text for details.

from which the impact to the total flux is minimal. To examine how the airborne EC system performed for the entire frequency spectrum, a Fast-Fourier-Transformation analysis has to be used.

5.3. EC performance test II: FFT analysis

Fast-Fourier-Transformation analysis was used as the quality checking tool for the collected data. The analysis was performed on 10 Hz data to examine fast-sampled aerosol concentration data, electrical noise in analog signals, etc., for every horizontal flight leg. Although results demonstrated considerable variability among transects, general characteristics emerged. Fig. 7 presents a sample of data from a portion of September 8th, 2003 flight: time series of vertical wind speed, air and sea surface temperatures, and aerosol concentration are shown in panels a, b and c, respectively. Corresponding power spectra are in panels d, e and f. Co-spectra of vertical wind speed with temperature and aerosol concentration are in panels g and h, respectively. The frequency was normalized by the ratio of averaged true airspeed and measurement height. Panel j highlights the 200-s portion of the flight (33 m elevation) from which data were selected for the analysis. The straight lines in panels d to h are theoretical slopes from similarity laws for the inertial sub-range, which are -2/3 for the power spectrum and -4/3 for the co-spectrum.

During the presented episode, the atmosphere close to the surface was unstable, with mixed MBL height reaching 550 m (spiral G in Fig. 5a). Sea surface temperature was higher than air temperature at 30 m altitude by 1.5 K. Particles were uniformly mixed from the measurement altitude to the top of mixed MBL (see spiral at G in Fig. 5d). Nevertheless, low frequency changes in aerosol concentration time series are noticeable in Fig. 7c.

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The shape of the vertical wind speed power spectrum multiplied by frequency shows energy decay for lower frequencies. The spectrum demonstrates the expected shape with a maximum at 0.1-1 Hz and a decay slope of -2/3 in the inertial sub-range, Fig. 7d. Similar shapes of vertical wind speed power spectra are described in Kaimal and Finnigan (1994) for the unstable surface and mixed layers.

The temperature power spectrum occasionally exhibited a white noise with the slope of +1 at normalized frequencies higher than about 1.5 Hz. It was due to the low level temperature variability in the marine boundary layer compared with an electrical noise generated in signal cables, i.e. low-signal-to-noise ratio. During the episode presented in Fig. 7e, this was a negligible white noise. The high end of the temperature power spectrum shown has only a slightly lower decay compared with the -2/3 slope. However, the noise is more visible in the power spectrum of other episodes. An experimental test confirmed that the noise was generated in the aircraft electrical cables extending from the airplane nose to the data logging system inside the cabin.

The vertical wind speed/temperature co-spectrum (Fig. 7g) was dominated by positive values due to the sea surface temperature being higher than the air temperature (Fig. 7b) during this transect. The decay at higher frequencies indicates that the aforementioned noise is not correlated with the vertical wind speed and therefore is not influencing flux estimates. The vertical wind speed and aerosol concentration co-spectrum (Fig. 7h) formed a "hill" shape with a decay slope at higher frequencies along the similarity law. The decay at lower frequencies indicates a sufficient length of time series used for the covariance calculations.

Both co-spectra have values of opposite sign to the total flux value for the same time interval. This could be explained by a plausible scenario in which air parcels rising to the sampling elevation may have previously descended from aloft. For the atmospheric stability being between neutral and weakly unstable, not all descending air parcels from the measurement height (about 33 m) towards sea surface necessarily reach the surface where heat exchange from the ocean surface occurs or aerosols are deposited. Some of them, at an intermediate height, may change direction and start to ascend again, bringing air to the measurement altitude. At that moment, vertical wind direction would be detected as being upwards; however, physical properties of the parcel are those of air from aloft. This would cause opposite signs to appear in the co-spectra. However, at the high frequencies this is probably due to random statistical error in spectral values, since the presented plot corresponds to one spectrum and not an average of many individual spectra. Aerosol power spectrum and co-spectrum presented in Fig. 7 are obtained from the measured aerosol concentration prior to desmearing or speeding-up. After the correction, the aerosol power spectrum increased by an order of magnitude at normalized frequency of around 1 Hz, and slightly increased or remained unchanged at low frequencies (not shown). The corresponding co-spectrum showed a significant increase in energy at higher frequencies and negligible increase at low frequencies (not shown).

"Hill"-type shapes and decays according to the -2/3 and -4/3 slopes in the presented power spectra and co-spectra are typically used as the quality criteria in EC studies. Results here confirm that the EC system implemented on the airborne platform is applicable for vertical flux measurements.

5.4. Aerosol number spatial variability

Examination of the spatial variability in particle concentration (Fig. 8) suggested that there is a major plume located almost parallel to the -122.2° longitude. The plume was crossed numerous times during the horizontal transects. Most likely, the plume originated from a ship motoring slightly west of the longitude. Close to the shore, where urban plumes reached offshore areas, the number concentration peaked at 20,000 particles cm⁻³ due to harbor and urban traffic particulates emission. The range of total number concentration at offshore locations and outside the plume throughout the flight was 500–1500 particles cm⁻³. Between the plume and the shore (at about -122° long), a number of short-duration plumes could be identified. Fresh plumes often exhibited a delta function-type jump in concentration times series, with numbers increasing to several tens of thousands particles cm⁻³. The increase typically lasted for several seconds to several tens of seconds, causing a low frequency distortion in the aerosol power spectrum. CPC 3010 and 3025 showed a good agreement when sub-20 nm particles were not present. Total aerosol number calculated from the size distribution data was in agreement with both CPCs. Occasionally, higher numbers were reported by CPC 3025 when nucleation mode aerosols were present due to the lower cut-off size of the instrument.

High aerosol concentration and variance indicated non-stationary conditions in which aerosol concentration fluctuations from mean values are higher or have different time scales than those corresponding to turbulent mixing. Episodes of stationary and low aerosol concentration representing clean marine aerosol are very preferable for testing airborne



Fig. 8. Spatial variability of the aerosol number concentration on September 8, 2003. The continent is East of the thick line.

aerosol flux measurements. However, due to many ship tracks, concentration time series that are stationary (for at least 3 min) were rarely observed. Low aerosol numbers typical of marine environment where the majority of the particles are sea salt (100–500 particles cm⁻³) were not detected. Coastal aerosol was significantly influenced by ship emissions and continental aerosol.

5.5. Aerosol size distribution: Marine case

In marine coastal environment, marine aerosol is superimposed by anthropogenic emissions from ships (Hobbs et al., 2000; Russell et al., 1999), which significantly increase Aitken mode aerosol concentration and frequently dominate the total particle number. Observed aerosol size distributions in this study are similar to the ones reported by Russell et al. (1999) from MAST experiment. According to their classification, observations from this study fall into the continentally influenced category, where the total aerosol number close to the sea surface outside of ship plumes is larger than 500 particles cm⁻³. In their study, chemical aerosol composition was comprised of equal fractions of ammonium sulfate, ammonium bisulfate, elemental carbon, and organic carbon related to emissions from diesel engines (25% each). Ship track aerosol significantly increased Aitken mode concentrations. Russell and coworkers presented measurements sampled in southwest direction by a 1° longitude from the flight tracks of this study. Their data indicated a negligible fraction of sea salt particles in total aerosol number.

Aerosol size distributions sampled at the most off-shore location during the case study flight are presented in Fig. 9. These data were sampled while the airplane transected from slightly northwest of point F towards G in Fig. 2. The figure shows measured dry and humid size distributions as well as two hypothetical humid size distributions. The hypothetical curves were obtained by shifting particle diameters in the dry distribution by certain growth factors to test whether the particles predominantly consisted of sea salt or sulfates. In the following analysis, it is assumed that hygroscopic response of sea salt particles to change in relative humidity is the same as that for sodium chloride particle. The growth factors of ammonium sulfate or sodium chloride at the RH used in the humid DMA (74% in this period) are 1.40 and 1.88, respectively. The similarity between the hypothetical (based on a growth factor of 1.40) and the humid aerosol size distribution strongly suggests a possibility of particles being composed mostly of ammonium sulfate. Fig. 9 indicates that the humid distribution can be approximately reconstructed from the dry one assuming ammonium sulfate particles



Fig. 9. A sample of surface aerosol size distributions from the segment between flight points D and F in Fig. 2 on September 8, 2003. Dry, wet, and two modeled growth factors of 1.4 and 1.88 are presented.

internally mixed with a negligible fraction of insoluble material. This applies for both the Aitken and accumulation mode aerosols. If the growth factor of 1.88 is used, the hypothetical distribution is significantly shifted to the larger sizes compared with the observed humid one.

A possibility remains that a small number of sea salt particles could be externally mixed with ammonium sulfate particles comprising a bulk part of entire aerosol population. Since the approach to estimate aerosol water uptake is not a single particle technique, it is insensitive to small amounts of sea salt. To address this, a sensitivity analysis was performed, where hypothetical wet size distributions were modeled, but in this case the initial dry aerosol was assumed to be an external mixture with 10%, 25%, or 50% of the particle number being NaCl and the remaining particles being ammonium sulfate (the corresponding 0 and 100% cases are presented in Fig. 9). For all three external mixtures, the hypothetical curves shifted towards larger sizes. For the 10% NaCl case, the hypothetical curve was very close to the pure ammonium sulfate curve—the difference was within the measurement uncertainty. However, for the 25% NaCl mixture, the difference between the hypothetical and wet size distributions was significant for aerosols smaller than 150 nm. For particles larger than 150 nm, the mixing ratio of NaCl that produces a shift that is readily distinguishable from the observed wet distribution is 50%. The increase in upper bound mixing ratio from 25% is due to the flatter size distribution with multiple small peaks for the larger size range (see Fig. 9) compared with the large uniform peak for the 10–150 nm size range. Thus, it can be concluded that 25% (and 50% for aerosols larger than 150 nm) is the upper bound for a possible externally mixed fraction of sea salt in the aerosol observed. It is likely that the actual fraction was less than that.

Sea salt in marine aerosol is generally distributed in the accumulation mode (approximately $0.1-1 \mu m$) and coarse mode (above $1 \mu m$) (Shinozuka, Clarke, Howell, Kapustin, & Huebert, 2004). In the September 8, 2003 flight, accumulation mode aerosols comprised 5–40% of the total aerosol number concentration. Since the estimated maximum fraction of NaCl particles is 50% of those larger than 150 nm and 25% for 100–150 nm aerosols, the maximum NaCl particle concentration in the analyzed areas, calculated from the measured size distributions, is 7% of the total aerosol number concentration. The range of 5–15% is the maximum possible NaCl particle number fraction for the entire flight. In practice, this number is likely to be smaller. Since the observed total aerosol concentrations range between 500 and 1500 particles cm⁻³, 7% equates to 35–105 particles cm⁻³. This concentration is very low and insufficient to significantly alter the total particle flux.

An important aspect of data interpretation is to determine if the aerosol is locally emitted (within the flux measurement footprint area) or advected from upwind sources. Locally emitted aerosol could be sea-salt particles or organic aerosol in Aitken mode via bubble burst mechanism, as recently proposed by O'Dowd et al. (2004). According to the findings in this section, the number of wind-driven emitted sub-micron sea salt particles is negligible. As reported by Russell

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Fig. 10. A sample of EC flux time series and corresponding aerosol total number concentration in top panel.

et al. (1999) from their measurements in Monterey Bay, Aitken mode aerosol chemical composition was similar to that of anthropogenic aerosols. In contrast, O'Dowd reported significantly higher relative organic carbon concentration in the aerosol in north Atlantic region. Sulfate particles are associated with anthropogenic emissions, and could be emitted locally from ships as described in the previous section. The internal mixture of sea salt and soot would be formed by coagulation; however, such process is slow and takes places over a time period of hours to days. During such a long time period, sea salt particles emitted from the ocean are mixed through the entire boundary layer and can no longer be treated as surface source particles. During this study, the internally mixed aerosol in coastal environment is advected to the footprint areas from upwind sources unless the airplane flew directly above a ship wake. At the shore line, a significant amount of sea salt is generated in the surf zone (de Leeuw, Neele, Hill, Smith, & Vignati, 2000), but this would be occurring downwind of the measurement area and is therefore irrelevant to the current study.

5.6. Vertical aerosol fluxes

Particle fluxes were calculated using 200-s time periods. Analysis showed that the flux was not significantly different if a 300-s time series was used; therefore, the shorter time period was chosen. However, for all particle fluxes, a 100-s time series yielded different values than 200-s time series, due to large low frequency variability in concentration time series caused by numerous ship emissions. Generally, measured fluxes exhibited a high variability due to ship plumes. Depending on the plume altitude relative to the aircraft's, the measured flux indicated either "emission" upwards or "deposition" downwards.

To demonstrate the time evolution of the measured flux, a segment of data from the "L"-shaped flight pattern D-F-G (Fig. 2) was chosen. The sampled aerosol was characterized with high number variability (Fig. 8) for most of the leg, and during the last several minutes, particle number behavior resembled background marine aerosol with continental influence. Fig. 10 demonstrates this transition. In the beginning of the segment at 21.17 UTC (Fig. 10), the airplane was about to exit a plume with maximum recorded aerosol number concentration of 2100 particles cm⁻³. The airplane was out of the plume by about 21.30 UTC where aerosol number dropped to below 1400 particles cm⁻³, and measured fluxes demonstrated a downward-directed transport. Data from that region of the Fig. 10 was used in Furje analysis above. More detailed analyses of aerosol fluxes related to plumes will be discussed in a separate manuscript. EC fluxes were increased by 30% prior to plotting in Fig. 10 to include Horst correction (black line). Two additional flux datasets are from the same raw data, but the concentration data were desmeared according to Collins et al. (2002) (red line) or speeded-up according to Kalogiros and Helmis (1993) (blue line) prior to covariance calculations. The deconvolution changed flux values significantly during periods when fluxes were in the order of $10^6 \text{ m}^{-2} \text{ s}^{-1}$ or smaller. When fluxes are higher (at about 21.18 UTC time in Fig. 10) the difference among the methods is minor. Two aerosol deposition episodes when aerosol number was not affected directly by a ship plume are analyzed below.

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5.7. Modeling deposition fluxes

It is interesting to find out how measured fluxes compare to aerosol flux determined by other than EC method. Aerosol dry deposition to water can be modeled using available models. In order to identify episodes with measured fluxes representing dry deposition, a rigid flux data filtering was performed. The following criteria were employed:

(1) Eliminate data segments sampled from higher than 50 m altitude and when standard deviation of the altitude was higher than 5 m over 200 s period. This limits data to constant-altitude legs.

(2) To avoid coastal influences and numerous spikes in concentration, data sampled east of -122.4° longitude were discarded.

(3) Only unstable atmospheric conditions were considered when selecting data from episodes in which heat flux was upwards and higher than 2 W m^{-2} .

(4) To avoid significant concentration spikes in the most northern flight segment (Fig. 8), only data south of 37.1° latitude were used.

(5) To avoid non-stationary episodes in aerosol concentration time series, only episodes with the SN/MN < 0.1 were used, where SN is the for standard deviation of aerosol number concentration within 200 s time period and MN represents an averaged concentration within the same time period.

Filtered concentration time series were visually examined, and all remaining unfiltered non-stationary episodes were removed manually. That procedure left seven legs longer than 200 s from the 4.5-h long flight. Five of them were close to the areas in the bay where aerosol number was not well mixed in vertical direction according to Fig. 5e. Flux data in those episodes represents vertical dilution rather than dry deposition, and they were removed. The remaining two episodes are summarized in Table 2. Their geographical locations are marked in Fig. 8. Presented values in the table are mean values for the each episode, plus or minus one standard deviation from the mean.

Fluxes were modeled using dry and wet aerosol size distributions. Removal of particles by water surface occurs due to Brownian diffusion, interception, and gravitational settling after turbulent mixing transports aerosol close to the surface and only a minimal distance in a laminar sub-layer remain between the particle and the surface. Slinn and Slinn (1980) modeled aerosol dry deposition by analyzing separately the turbulent transport from the measurement altitude to the sub-layer and the transport across the sub-layer and assuming a constant net flux versus height. Modeled dry deposition velocity is a function of particle size, which for hygroscopic particles depends on ambient relative humidity. The uncertainty in particle size changes close to the water where RH is elevated to about 98% challenged similar studies in the past. In our case, ambient RH at measurement altitude was about 80% on that day. As discussed above, it is reasonable to assume ammonium sulfate aerosol composition with respect to water uptake properties. Using wet or dry aerosol size distribution, we apply the Slinn model:

$$F_{\text{Slinn}} = \int_{d_{\text{p}}^{\min}}^{d_{\text{p}}^{\max}} c_{d_{\text{p}}} \cdot v_{d_{\text{p}}} \delta d_{\text{p}},$$

where $d_p^{\min} = 10 \text{ nm}$ and $d_p^{\max} = 850 \text{ nm}$, the range of measured aerosol size distribution, c_{d_p} is the aerosol number density function from the measured aerosol size distribution, d_p is the particle diameter, v_{d_p} is the dry deposition velocity of a particle of size d_p derived from Slinn model using measured meteorological data. In this manner, a total deposition flux can be calculated (Buzorius et al., 2003). The values of c_{d_p} were interpolated to 1-nm resolution from the measured aerosol size distribution, and v_{d_p} was calculated with 1-nm steps as well. For the "wet" case, the dry distribution, c_{d_p} , was shifted by a growth factor of 2 to represent ambient aerosol close to the water surface where relative humidity is close to 98%. The assumed growth factor is slightly smaller compared with the bulk values for the ammonium sulfate. The reduction occurs due to the surface tension effect on sub-100 nm particles. If no other aerosol sinks are present, the measured deposition fluxes should agree with the calculated values assuming that the model is valid.

Modeled fluxes yielded lower vertical transport intensity in absolute values compared with the measurement (Table 2). Both deconvolution methods significantly increased downward fluxes during event #1 and decreased them during event #2 (the location of each event is shown in Fig. 8). Higher fluxes during the event #1 were probably due to aerosol plume aloft (see Fig. 5e) slightly north, and the measured flux probably represented local flux. Slinn wet and dry deposition fluxes have very similar values. The measurement uncertainty due to CPC mixing time is eliminated in desmeared and speeded-up fluxes. The random error due to uncertainty in vertical wind speed measurements was about 0.12×10^6 particles cm⁻² s⁻¹. The error due to counting statistics was higher, reaching 0.21×10^6 particles cm⁻² s⁻¹ in event

Table 2 Measured (E concentration	C) and modeled	d fluxes during ation was calcu	two deposition lated prior to th	episodes. Errors te de-convolution	s estimates and n	d mean charac	teristics present	ted. EC fluxes	were increased	1 by 30% to acc	ount for Horst cor	rection. Particle
									Standard de	viation	Error in EC flux	
Nr. of episode	Flux, EC (×10 ⁶ # $m^{-2} s^{-1}$)	Flux, de-smear $(\times 10^6 \#$ $m^{-2} s^{-1})$	Flux, speed-up $(\times 10^6 \ \mu$ $m^{-2} \ s^{-1})$	Flux, Slinn wet $(\times 10^6 \text{m}^{-2} \text{s}^{-1})$	Flux, Slinn, dry $(\times 10^6 \ \mu$ $m^{-2} \ s^{-1})$	Friction velocity $(m s^{-1})$	Hor. mean wind speed $(m s^{-1})$	Aerosol mean conc. $(\# \text{ cm}^{-3})$	Aerosol conc. (# cm ⁻³)	Vert. wind speed $(m s^{-1})$	error in $\delta w (\times 10^6 \text{ #}$ m ⁻² s ⁻¹)	Counting stat. $(\times 10^6 \text{ #} \text{ m}^{-2} \text{ s}^{-1})$
-	-0.3579	-0.82	-0.78	-0.2490	-0.2534	0.5	10.3	1130	25	0.28	0.1135	0.1146
5	± 0.0429 ± 0.0429	± 0.10 -0.28 ± 0.21	± 0.19 -0.16 ± 0.18	± 0.0394 -0.2580 ± 0.0383	± 0.0413	0.5	14.4	1500	50	0.49	$\pm 0.0000 \pm 0.0009 \pm 0.0009$	± 0.0006 ± 0.0006

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#2. The total counted number of particles per time interval is significantly increased using longer (300 s vs. 200 s) time intervals. An additional error due to the stochastic nature of turbulence is typically estimated to be 10% (Rannik & Vesala, 1999).

6. Conclusions

In this pilot study we presented airborne aerosol flux measurement system. Aerosol number concentration was measured with a condensation particle counter aboard the Twin Otter aircraft, and the particle fluxes were calculated using the eddy-correlation method. The technique provides valuable data on vertical aerosol fluxes and can be used for mapping aerosol surface sources and sinks on 10 km grid scale. The measurements were validated using FFT analysis and by comparing EC-derived and bulk measurements of the sensible heat flux. The EC system performed well for the entire frequency co-spectrum and the expected isotropic inertial subrange (rapid decrease) was observed at high frequencies.

Measured aerosol number concentrations varied from several hundreds to almost 30,000 particles cm⁻³. Large peaks in concentration were detected close to the shore/urban environment and close to ships. Measured particle fluxes were changing direction numerous times during the flight. Most of the large fluxes are due to aerosol plumes and are associated with boat emissions. Depending on the plume elevation relative to the aircraft altitude, the measured flux could be up or down. Anthropogenic particles dominated the aerosol size distribution. No two flights had similar flux spatial variability due to chaotic boat traffic, as flux signal was dominated by plume spikes. For more detailed analysis, a case study of the September 8, 2003 flight was chosen. On that day wind speeds were one of the largest during the study and dry and humid aerosol size distribution data were available. Comparison of "humid" and "dry" aerosol size distribution measurements revealed that outside the plume the aerosol hygroscopic growth factor was very similar to that of ammonium sulfate (1.4 at 74% RH). The number of NaCl particles in ambient aerosol was minimal with the upper bound of the fraction being 7%.

Local fluxes of particles at approximately 33 m elevation in coastal areas are primarily controlled by the concentration gradients and atmospheric mixing. Atmosphere mixing was primarily due to wind shear and was not buoyancy-driven, which possibly prolonged the existence of gradients. Mixed MBL height decreased as it got closer towards the shore, where wind speeds were reduced. Inside the bay, the reduction in the MBL height from 500 to less than 70 m was observed approaching the shore from west to east, as wind speeds dropped from 14 m s^{-1} to less than 6 m s^{-1} .

Aerosol deposition episodes were isolated from the entire dataset by applying strict data filtering criteria. During those episodes fluxes were directed downwards and had averaged values of $-(0.36 \pm 0.22) \times 10^6$ and $-(0.34 \pm 0.04) \times 10^6$ particles m⁻² s⁻¹ if the CPC slow response accounted for by a 30% increase in flux values. Corresponding modeled values showed slightly lower deposition rates, but the difference was within the measurement uncertainty. Those values were calculated using Slinn's model, measured aerosol size distribution, and measured meteorological parameters. Here, we restrain from validation of Slinn's model itself, due to very limited dry deposition dataset.

The major uncertainty sources in EC flux measurements were due to particle counting statistics, CPC slow response time, and error in vertical wind speed measurement. The error due to particle counting statistics was about $0.1-0.2 \times 10^6$ particles $m^{-2} s^{-1}$. If the total flux is small, this source of error may have a significant impact. The error can be reduced by increasing the time interval during which a covariance is calculated with the cost of reduced spatial resolution in flux data set. Sampling in areas with the higher aerosol number results in a smaller uncertainty. Flux measurement uncertainty due to errors in the vertical wind speed determination was about 0.1×10^6 particles m⁻² s⁻¹. We note that the flux error due to the slow CPC response is a systematic error, which can be corrected by either assuming a surface similarity theoretical form for the corresponding co-spectrum or directly correcting the time series of CPC concentration measurement. In the latter case, the error after the correction is negligible. The flux loss due to small sampling frequency (10 Hz) for the given airspeed (50 m s⁻¹) and measurement altitude (about 33 m) is negligible for the usually unstable and nearly neutral atmospheric conditions observed in the MBL when fast sensors are used. For the slow sensor such as CPC, the flux loss can be corrected by systemic corrections, and three methods (spectral, desmearing and speeding-up) were presented. Finally, for the 200-s averaging time interval used in this study, the flux error due to the stochastic nature of turbulence was about 10%. Concluding, the uncertainty in airborne aerosol flux measurements is about $0.1-0.3 \times 10^6$ particles cm⁻² s⁻¹. The uncertainty decreases with increasing total aerosol number or increasing time periods for which aerosol flux is calculated.

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