Direct Measurements of New-Particle Fluxes in the Coastal Environment

Robert J. Flanagan, Michael Geever, and Colin D. O'Dowd

A Department of Experimental Physics and Environmental Change Institute, National University of Ireland, Galway, University Road, Galway, Ireland.
B Corresponding author. Email: robert.flanagan@nuigalway.ie

Environmental Context. The formation of new secondary aerosol particles in the natural atmosphere is important in terms of controlling the background aerosol population, which significantly impacts on climate. The coastal zone is perhaps the strongest natural source of new secondary aerosol particles, driven by the release of biogenic vapours, which, after undergoing photochemical reactions, lead to the massive production of nucleation mode aerosols, with concentrations often reaching in excess of 10^6 cm^{-3}. Quantification of this source strength is important, particularly on a regional scale, in terms of estimating the impact of aerosols on climate.

Abstract. Measurements of the flux of new secondary aerosol particles during nucleation events in the coastal environment using an eddy-correlation technique are reported for the first time. Events are divided into two types based on the prevailing wind direction. During tidal-related nucleation events, new-particle upward fluxes are typically of the order of 10^9–10^{10} particles m^{-2} s^{-1}. A close correlation (r^2 = 0.86) was found between total particle concentration and total (positive) flux when air masses were not affected by multiple sources. This would suggest that continuous measurements of particle number concentration at Mace Head can be translated into a flux using the resulting parameterization. It is expected that parameterizations obtained from similar data and analysis would be equally feasible at other coastal locations.

Keywords. aerosol fluxes — aerosol nucleation — climate change — coastal aerosols

New-particle production resulting from the nucleation of stable clusters from the gas phase and their subsequent growth, both driven by the rapid production of condensable iodine vapours, is observed to readily occur in the coastal environment. The rapid production of iodine vapours occurs when macroalgae in the tidal zone is exposed during low tide. The algae release both inorganic and organic iodine vapours, particularly in the form of I_2 and CH_3I, both of which undergo rapid photolysis leading to the production of condensable iodine oxides. The current state-of-the-art relating to iodine chemistry and new-particle production is summarized in other papers in this issue. Coastal nucleation has been studied most extensively at the Mace Head research station (53°20′N, 9°54′W) on the Irish Atlantic coastline; however, similar nucleation events have been observed on the Scottish coast as far back as 1897 and also more recently. A full characterization of nucleation events at Mace Head can be found in O’Dowd et al. The events can be characterized as the massive production of nanometer-sized particles lasting for periods of 1–8 h depending on tidal amplitude and meteorological variables. The events can occur in both polluted and clean air. Under clean air conditions, particle concentrations can increase from the background level of around 500 cm^{-3} to in excess of 10^6 cm^{-3}. This study aims at quantifying the flux of these new particles in the coastal environment through the measurement of turbulent fluxes using an eddy-correlation (EC) technique.

Aerosol particle fluxes have been measured in a number of locations using the EC method but these are the first such measurements of secondary aerosols during coastal new-particle formation events. Given the frequency of these events and the very large concentrations that can be produced (10^6 cm^{-3}), it is important to quantify the flux of these particles into the atmosphere. The flux measurements were performed at Mace Head over a 7-week period during August and September 2002. A standard EC package incorporating a Gill Windmaster Pro Solent sonic anemometer (Gill Instruments Ltd, Lymington, England) to provide 3D wind fields at 10 Hz and a TSI Condensation Particle Counter (CPC) model 3025 (TSI Incorporated, Minnesota, USA) with a low cutoff of 3 nm were used. A more detailed description of the flux package and its use (for primary marine aerosol fluxes) can be...
Although the peak particle concentration was similar for both background levels of less than 1000 cm$^{-3}$, the flux of particles during this case was mostly negative with a peak (negative) value of $-2.77 \times 10^{10}$ m$^{-2}$ s$^{-1}$. For part of the event, there was a positive flux peaking at $0.24 \times 10^{10}$ m$^{-2}$ s$^{-1}$. Similar characteristics were observed for the other nucleation events during the measurement period. The magnitude of these fluxes compare very well with those calculated using chamber experiments.$^{[14]}$ They reported a flux of $3 \sim 3.4$ nm particles of $2.5 \times 10^{10}$ m$^{-2}$ s$^{-1}$ for a seaweed loading of $2.5$ kg m$^{-2}$, which is representative of a typical seaweed density at Mace Head. It should be noted that these fluxes are over three orders of magnitude greater than primary marine aerosol fluxes.$^{[10]}$

The difference in aerosol flux characteristics for the two event types is a result of their differing footprints. For a Type I event, there is only one source in the measurement footprint and that is approximately 50–100 m from the measurement point. Since the measurement point is directly in the local coastal plume and close to the plume head, the new particles have not yet become well mixed in the boundary layer. By comparison, for the Type II event, there is still the local coastal plume, but now also multiple strong sources upwind. These multiple sources are sufficiently far upwind to allow the newly produced aerosols to become well mixed in the boundary layer and, consequently, the new downward flux of the mixed particles more often than not dominates the upward flux of the local coastal plume. Thus, these measurements indicate that the quantification of new-particle source fluxes is only achievable under Type I conditions. It should be noted that we are measuring the flux at a 10 m height and not the surface source flux. A minor degree of mixing could occur between the source region and the measurement setup, which would lead to an underestimation of the source flux. In order to obtain a surface source flux, a footprint model specific to Mace Head would need to be employed. This, however, is beyond the scope of the current work.

Continuous measurements and analysis of coastal aerosol fluxes is very resource demanding and is not performed on a continuous basis. However, it was found that the particle flux and particle concentration were strongly correlated for Type I events (see Fig. 3). The relationship can be described by:

$$\log_{10} F = m \log_{10} N - k$$

where $N$ is the particle concentration in number per cm$^3$ and $F$ is the particle flux in number $\times 10^6$ m$^{-2}$ s$^{-1}$ with a slope of $m = 1.40 \pm 0.16$, an intercept of $k = -3.46 \pm 0.65$ and a correlation coefficient of $r^2 = 0.86$. A similar strong correlation did not exist for Type II events. Using the above relationship, it is suggested that data from the continuous monitoring of particle concentration at Mace Head can be readily translated into a particle flux with an acceptable degree of accuracy. We expect that parameterizations obtained from similar data and analysis would be equally feasible at other coastal locations.

In summary, eddy-correlation fluxes of nucleation mode secondary aerosol particles produced in the coastal environment have been quantified for the first time. Near the particle source region, the particle flux is upward and can reach the order of $10^9 \sim 10^{10}$ particles m$^{-2}$ s$^{-1}$ during an event. The magnitude of these fluxes is in excellent agreement with those derived from chamber experiments. Upward fluxes are positively correlated with particle concentration under conditions

---

**Fig. 1.** A typical nucleation event at Mace Head observed during the measurement period. Total particle concentration (1 Hz data) is for sizes larger than 3 nm (dark grey line). Tidal amplitude for that day is also included (light grey line).

---

**Table 1.** The difference in aerosol flux characteristics for the two event types is a result of their differing footprints. For a Type I event, there is only one source in the measurement footprint and that is approximately 50–100 m from the measurement point. Since the measurement point is directly in the local coastal plume and close to the plume head, the new particles have not yet become well mixed in the boundary layer. By comparison, for the Type II event, there is still the local coastal plume, but now also multiple strong sources upwind. These multiple sources are sufficiently far upwind to allow the newly produced aerosols to become well mixed in the boundary layer and, consequently, the new downward flux of the mixed particles more often than not dominates the upward flux of the local coastal plume. Thus, these measurements indicate that the quantification of new-particle source fluxes is only achievable under Type I conditions. It should be noted that we are measuring the flux at a 10 m height and not the surface source flux. A minor degree of mixing could occur between the source region and the measurement setup, which would lead to an underestimation of the source flux. In order to obtain a surface source flux, a footprint model specific to Mace Head would need to be employed. This, however, is beyond the scope of the current work.

Continuous measurements and analysis of coastal aerosol fluxes is very resource demanding and is not performed on a continuous basis. However, it was found that the particle flux and particle concentration were strongly correlated for Type I events (see Fig. 3). The relationship can be described by:

$$\log_{10} F = m \log_{10} N - k$$

where $N$ is the particle concentration in number per cm$^3$ and $F$ is the particle flux in number $\times 10^6$ m$^{-2}$ s$^{-1}$ with a slope of $m = 1.40 \pm 0.16$, an intercept of $k = -3.46 \pm 0.65$ and a correlation coefficient of $r^2 = 0.86$. A similar strong correlation did not exist for Type II events. Using the above relationship, it is suggested that data from the continuous monitoring of particle concentration at Mace Head can be readily translated into a particle flux with an acceptable degree of accuracy. We expect that parameterizations obtained from similar data and analysis would be equally feasible at other coastal locations.

In summary, eddy-correlation fluxes of nucleation mode secondary aerosol particles produced in the coastal environment have been quantified for the first time. Near the particle source region, the particle flux is upward and can reach the order of $10^9 \sim 10^{10}$ particles m$^{-2}$ s$^{-1}$ during an event. The magnitude of these fluxes is in excellent agreement with those derived from chamber experiments. Upward fluxes are positively correlated with particle concentration under conditions
Fig. 2. (a) Half-hour averages of total particle flux (black line), particle concentration (grey dashed), wind direction (circles) for a Type I event. Tidal height for that day is also included (grey line). (b) Half-hour averages of total particle flux (black line), particle concentration (grey dashed), wind direction (circles) for a Type II event. Tidal height for that day is also included (grey line).

Fig. 3. Relationship between half-hour averages of particle concentration and vertical particle flux for Type I nucleation events in the coastal environment. Included are the linear regression fit and 99% confidence (dashed) and prediction (dotted) intervals.

where the air has advected over one source region near to the sampling point (Type I events) and suggests that under these conditions, the particle concentration can be used as a surrogate for particle fluxes in the absence of direct flux measurements. Fluxes during nucleation events were found to be over three orders of magnitude greater than primary marine aerosol fluxes and can provide a significant source of new aerosol particles on a regional scale.

Acknowledgements

This work was supported by Enterprise Ireland under contract SC/2001/200, the European Commission under contract EVK-CT-2001-00127 (QUEST) and the Irish Environmental Protection Agency.

References

Direct Measurements of New-Particle Fluxes in the Coastal Environment


