Dear editor,

Thanks for your effort in organizing the review of this paper. Please see our replies to referees' comments below, which we have kept in *Italic*. The track change version of the manuscript is copied at the end.

Referee#1: This paper is a description of an analysis of aerosol measurements made at Penlee Pt (PPAO) on the coast of the UK. Eddy correlation aerosol fluxes are computed and are interpreted as the sum of source and deposition fluxes. An effective aerosol source strength is computed and analyzed with considerations of wind direction, etc.. The authors find the source strength correlations better with wave height and/or wave parameters than with wind speed. The source is stronger than expected for open ocean but weaker than that observed directly from a surf zone. The interpretation is that wave breaking is more intense (or something) in a shallow zone close to shore than the open ocean. Most of the number flux occurs for aerosols greater than .1 and less than 1 micron. The paper is well written and the authors have carefully considered a number of experimental and physical aspects of the analysis and interpretation. In my view it can be published in its present form. I have a few comments the authors may with to consider.

*I suggest the authors make it painfully clear that their results are not affected by surf generation and the enhanced production they see is associated with enhanced breaking in shallow water but external to the shore break. Maybe they thought it was obvious but I pondered this.

Thanks for the suggestion. We did state this in the paragraph beginning at line 171 already but will make this point clearer. We will add in the abstract and the conclusion a sentence to the effect of "Sea spray fluxes measured from PPAO came essentially all from the shallow waters, and were not noticeably affected by the shore break/surf zone."

*I suggest they carefully check terminology of net, source, and total aerosols. I found myself wondering if they were consistent. A number of figures say 'total aerosol number flux' but I am not confident I know if it is net or source.

Thanks for the suggestion. We will make it clearer in the revision that 'total' refers to either the CPC flux or the CLASP flux integrated over all size range; 'source' indicates total flux that has been corrected for deposition.

Line 91. I don't think the turbulent flux is the same as the net flux. To me, net=turb-VgC, where Vg is the gravitational fall velocity.
Net=Source-VdC=turb-VgC
For the sizes they are considering, it may be that Vg is much less than Vd. They should state this. If Vg is negligible, then Source=turb+VdC

Sorry for not explaining this more clearly in the original text. Our calculation of the aerosol deposition velocity accounts for size-dependent gravitational settling already. This will be made obvious in Section 2.

On line 94 they state that source is obtained by subtracting deposition from net Source=turb-VdC
Doesn't seem consistent with Fig 5, where source is greater than net. Please check this and get it straight.

This is because we adopt the sign convention of Vd being negative. We will make this clearer in Section 2 and through out.

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Thanks for the suggested literature. They are consistent with our assessment that aerosol fluxes and concentrations are not in equilibrium at PPAO.

Figure 11 is certainly interesting. It is surprising that aerosol spectral concentration and source flux is independent of wind speed. The graphs might be a little easier to use of the vertical axis was multiplied by R (are conserving).

The aerosol source flux distribution is not quite independent of wind speed: fluxes of small aerosols are slightly higher at higher wind speeds (as indicated by the error bars), but the bin-separation is not nearly as clear as by wave height. We prefer to keep the vertical axes in dN/dR80 and dF/dR80 as they are the convention in literature.

Referee#2: This contribution describes total and size resolved Eddy Covariance measurements of aerosol number flux at a coastal research station. The total number flux measured with a CPC integrates particles larger than 1.5 nm. While size resolved fluxes between 0.1 and 6 um are derived from a CLASP. The authors combine the measured net fluxes with size resolved dry deposition velocities derived from the model of Slinn and Slinn (1980), in order to obtain the source flux over the footprint area. The dependence of the size resolved and size-integrated source flux on wind speed, wave height, and wave Reynolds number is studied in detail and compared with previous parametrizations that where derived over the open ocean and in coastal waters. The authors conclude that their measurements and findings are representative for coastal waters (excluding the surf zone) where sea spray emissions are largely enhanced due to costal wave breaking. Eddy Covariance measurements of aerosol fluxes in the marine environment provide an almost direct way to measure sea spray emission fluxes, however due to the complexity of making these measurements only few data sets exist today. Quantification of coastal sea spray emissions are relevant to the aerosol concentration and composition in the densely populated coastal regions and may thus bear relevance for heath related studies. Comparing the relation of coastal and open ocean sea spray emissions to forcing parameters may help to further improve the understanding of sea spray emission in general. I therefore consider this paper to be a valuable contribution and recommend publication with minor revisions as outlined below.

*As stated by referee Christopher Fairall the source flux can be derived from the mea- sured turbulent flux (FEC) via Source = FEC+Vd*C-Vg*C, where C is the particle con- centration, and Vd and Vg are the dry deposition and the settling velocity respectively. Vg depends on the particle size, shape and density, and hence on the relative humidity and the hygroscopy of the particles.

Our calculation of the aerosol deposition velocity accounts for size-dependent gravitational settling already. We have made this clearer in the revision (especially section 2).

The deposition velocity Vd may in additional be affected by turbulent and diffusive processes. Various dry deposition parametrizations can be found in the literature. The authors settle for the model of Slinn and Slinn (1980), which assumes that the particles assume their dry diameter in the constant flux layer and grow to a diameter defined by the growth rate and the assumed equilibrium relative humidity above seawater. Depending on the ambient relative humidity, the assumption of dry sea salt particles in the constant flux layer may introduce some errors. I therefore recommend the authors to explicitly state how they apply Slinn and Slinn (1980), i.e. which diameter conversions are performed, what particle densities are assumed.

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We compute the aerosol deposition velocity (accounting for gravitational settling) per Eq. 4 from Slinn and Slinn 1980.
Aerosol size change as a function of humidity is computed following Gerber (1985) and we assume a dry aerosol density of 2170 kg/m3 (that of sea salt).

*I further suggest that the ambient relative humidity time series should be presented in Fig 3., since it should play an important role in determining the correction term (Vd*C- Vg*C).*

A time series of relative humidity during this example period is shown below and is now added to Fig.3.
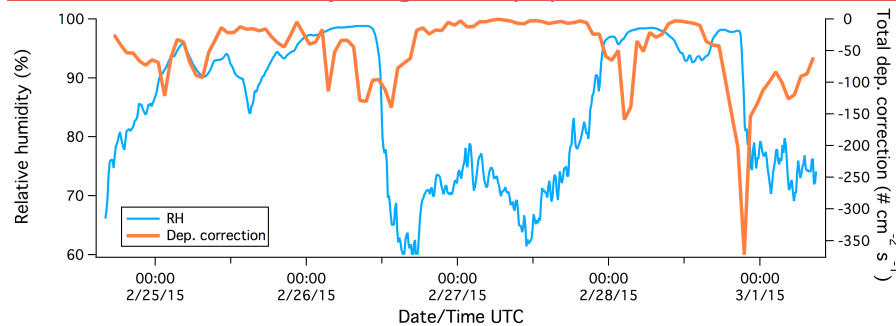
Line 103 to 105: Please consider splitting this sentence into two. Maybe the time series of the correction factors could be provided in the appendix to illustrate the variability of the relative importance of the correction term.

We have revised the sentence to: *"Integrated over all CLASP sizes, this deposition correction amounts to 25 cm^{-2} s^{-1} in the mean (up to ~200 cm^{-2} s^{-1}) when winds were from the southwest. This represents 14% of the net flux on average (up to ~50%)."* The total deposition correction term for the CLASP (including gravitational settling) is shown in the figure above, and we adopt the formula of source flux = net flux – deposition_correction.

Line 109: Please specify which of the formulations, provided in Gerber (1985), you are using and with which set of coefficients?

The formulations and coefficients (in Matlab) are copied below.
A=0.7674; B=3.079; C=2.572E-11; D= -1.424; % seasalt coefficients

% define dry radius space
Rd = [0.0001:0.0001:20]';

 % grow dry radii to ambient humidity RH1 (RH1 between 0 and 1)
 Ra = ((A*Rd.^B)./(C*Rd.^D - 0.434*log(RH1)) + Rd.^3).^(1/3);

 % find dry radii for radii measured at RH1
 for n=1:length(R1);
  k = max(find(Ra<R1(n)));
  R1d(n) = Rd(k)+diff(Rd(k:k+1))*((R1(n)-Ra(k))/diff(Ra(k:k+1)));
 end

% and calculate the radii grown back to required humidity RH2 (here 0.8, or 80%)
R2 = ((A*R1d.^B)./(C*R1d.^D - 0.434*log(RH2)) + R1d.^3).^(1/3);

Line 115: please correct "off of"
We have replaced 'off of' with 'from.'

Line 138: R (=radius) has not been defined previously. Theres is some chance of confusion with the coefficient of determination. At this note, please unify the use of italic and noraml font for variables like Hs and RHw.
We have chosen to spell out radius here and not use R to indicate radius. We have made sure that significant wave height and wave Reynolds number are thoroughly italicized in the revision.

Line 134 to 142: How much does Vd predicted by Slinn and Slinn (1980) vary over the range of conditions encountered during the observations? Would accounting for these variations reduce the scatter in Figures 4, 6, and 7? However, these variations may be small compared to the uncertainties introduced by the unknown size distribution below 0.1 um.
Vd varied from -0.007 to -0.13 cm/s during this period. The wind dependent Vd correction WAS applied to the CPC data, thus accounting for these variations already. We have revised the sentence to the following to be clearer: 'We compute the wind speed dependent V_d at these two aerosol sizes using the Slinn and Slinn (1980) parameterization, which amounts to -0.034 and -0.010 cm s^{-1} for the mean conditions at PPAO. For simplicity, we take the average of the two V_d datasets and multiply it by the CPC number concentration to estimate the deposition flux.'

Line 189 to 190: Replace "likely" with "obviously". What other difference is there be- tween the source and net fluxes than the deposition correction scheme?
We have changed the sentence to "The source fluxes show greater discrepancy due to the different deposition correction schemes applied."

Line 222: Please state the parameters used in the parametrization of Resio et al (2002).
We have revised the line to "Here H_s in the Plymouth Sound is predicted using a parameterization for fetch-limited waters (Resio et al. 2002) as a function of fetch and friction velocity since a direct measurement is not available."

Line 247 ". . . dependence of the sea spray flux . . ."
Suggestion accepted.

Line 265: May this be because above 2 um the deposition and settling velocities become so large that the correction term plays an too important role, thereby increasing the uncertainty of the source flux estimation?
Our statement was on the number distribution, and not on the fluxes.

Line 272 to 275: please specify to which size range the lognormal mode is fitted to? From the data there is no evidence that the peak of dF/dR80 is at R80=0.1um. I suggest that you also assume a lower mode center e.g. 50 nm or 70 nm, in order to estimate he uncertainty of the correction for the missing film drop flux.
The lognormal mode was allowed to be fitted from 10 nm to 7 micron radius, but in practice has a width of approximately 80 to 150 nm in our fit.
We refrain from making a lognormal fit centered at a lower mode (e.g. 50 or 70 nm) for two reasons: first, with less than half of the mode captured by the CLASP, the uncertainty of the fit would be very large; second, some previous measurements of marine aerosols often show a mode at a measured radius of 100 nm, hence our assumption that the lowest bin measured by the CLASP represents this mode size. Future

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measurements of finer aerosol size distributions (e.g. by a SMPS) at our site should provide more information on the robustness of our assumption.

Line 276: What is the reason for integrating from 10 nm rather than from 1.5 nm?
It makes no difference whether the log-normal fit is integrated from 10 nm or from 1.5 nm, as the fit is already at zero at 10 nm.

Line 275 to 278: What is the reason for comparing the median of the correction to the mean of the number flux difference? Considering the large variability of the total number fluxes (Fig 3. B), the median of the number flux difference may be the more robust estimate.
Good point. We have revised the sentence to "This is consistent with the finding that the total aerosol number flux from the CLASP was lower than that from the CPC by a mean (median) of 45 (40) $cm^{-2} s^{-1}$."

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Sea Spray Fluxes from the Southwest Coast of the United Kingdom –

Dependence on Wind Speed and Wave Height

240   Mingxi Yang[1], Sarah J. Norris[2], Thomas G. Bell[1], Ian M. Brooks[2]

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Abstract. Fluxes of sea spray aerosols were measured with the eddy covariance technique from the Penlee Point Atmospheric Observatory (PPAO) on the southwest coast of the United Kingdom over several months from 2015 to 2017. Two different fast-responding aerosol instruments were employed: an ultra-fine condensation particle counter (CPC) that detects aerosols with radius above ca. 1.5 nm, and a compact lightweight aerosol spectrometer probe (CLASP) that provides a size distribution

250   between ca. 0.1 and 6 μm. The measured sea spray emission fluxes essentially all originated from the shallow waters upwind, rather than from the surf zone/shore break. Fluxes from the CPC and from the CLASP (integrated over all sizes) were generally comparable, implying a reasonable closure in the aerosol number flux. Compared to most previous observations over the open ocean, at the same wind speed the mean sea spray number fluxes at PPAO are much greater. Significant wave height and wave Reynolds number explain more variability in sea spray fluxes than does wind speed, implying that enhanced wave breaking

255   resulting from shoaling in shallow coastal waters is a dominant control on sea spray emission. Comparisons between two different wind sectors (open water vs. fetch-limited Plymouth Sound) and between two sets of sea states (growing vs. falling seas) further confirm the importance of wave characteristics on sea spray fluxes. These results suggest that spatial variability in wave characteristics need to be taken into account in predictions of coastal sea spray productions and also aerosol loading.

260   **1. Introduction**

       Sea spray aerosols formed from wave breaking impact the Earth's radiative balance both directly by scattering light (Haywood et al. 1999; Lewis and Schwartz, 2004) and indirectly by affecting marine cloud formation (Clarke et al. 2006). At high wind speeds sea spray also has the potential to influence the air-sea transfer of heat and gases (e.g. Andreas et al. 1995; Jeong et al. 2012). From the perspective of atmospheric chemistry, sea spray droplets provide an important medium for

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heterogeneous reactions (Sievering et al. 1991; Kim et al. 2014). In coastal regions where roughly half of the world's population resides, sea spray constitutes a major component of particulate matter in the marine atmospheric boundary layer. Particulate matter is directly relevant for human health and is subject to air quality regulations.

Breaking waves entrain air into water, resulting in plumes of bubbles within the top meters of the ocean and the appearance of whitecaps (Thorpe, 1992). Sea spray aerosols are primarily formed from the bursting of those bubbles. Two modes of sea spray aerosols are commonly observed: the film drop mode (predominantly submicron) arising from disintegration of relatively large bubbles, and the jet drop mode ($\geq$500 nm to a few microns in radius) from relatively small bubbles (e.g. Blanchard, 1963). Tearing of the wave crest under conditions of very high winds can also lead to large spume droplets (tens of microns to millimeters; Monahan et al., 1983), which may contribute significantly to the total sea spray mass flux but not to the number flux. The causal relationship between bubbles and sea spray has led to the application of the "whitecap" method for the estimation of sea spray flux over the ocean: in which the spray production flux from unit area of a foam surface is prescribed based on other measurements and scaled up by the whitecap fraction (e.g. Monahan et al. 1986; Martensson et al. 2003; Clarke et al. 2006). However, the parameterization of whitecap fraction as a function of wind speed has an uncertainty of about an order of magnitude in moderate winds (e.g. Anguelova and Webster, 2006).

Recent works suggest that the whitecap fraction is also sensitive to parameters such as sea state (Scanlon and Ward, 2016; Brumer et al. 2017a) and water temperature (Salisbury et al. 2014; Salter et al. 2015). In shallow waters, wave breaking (and so whitecap fraction) depends not only on wind forcing, but also on the interactions between wind waves and swell with the bottom topography (e.g. de Leeuw et al. 2000; Resio et al. 2002). The bubble number concentrations within the surf zone can be about two orders of magnitude higher than over the open ocean (e.g. Brooks et al. 2009). van Eijk et al. (2011) estimated surf zone aerosol flux on sandy beaches to be about an order of magnitude higher than for the open ocean under similar wind conditions.

There are only a few datasets of direct measurements of sea spray fluxes by the eddy covariance (EC) method. Based on data from an Arctic cruise, Nilsson et al. (2001) published the first EC measurements of aerosol number fluxes, which correlated strongly with wind speed. Their data suggest that sea spray source flux consists of a film drop mode centered at ~50 nm radius and a jet drop mode center at 500 nm radius. Geever et al. (2005) measured submicron aerosol fluxes at radius > 5 nm and > 50 nm at the coastal station of Mace Head on the west coast of Ireland. They found comparable aerosol number fluxes in the Aitken mode (5–50 nm) and in the accumulation mode (50–500 nm). Norris et al. (2008) measured size-distributed aerosol fluxes between 0.15 and 3.5 μm radius at the Duck Pier on the east coast of United States. They showed that sea spray flux increases with the local wind speed up to a radius of 1 μm. More recently Norris et al. (2012, 2013b) measured size distributed sea spray fluxes (0.18 < radius < 6.61 μm) over the open ocean, and explored the wind speed and wave Reynolds number

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dependences of the flux. The wave Reynolds number (primarily a function of wind stress and significant wave height) was found to explain about twice as much variance in the open ocean sea spray fluxes than wind speed alone (Norris et al. 2013b).

Total and size distributed aerosol number fluxes have seldom been measured simultaneously, precluding an assessment of the sea spray flux closure and distribution. Here we report concurrent measurements of effectively "total" (radius > 1.5 nm) and size distributed sea spray fluxes (about 0.1 < radius < 6 μm radius) from a coastal site over several months from February 2015 to February 2017. We examine how sea spray fluxes vary with wind speed (1–21 m s^{-1}), significant wave height (0.2–3.1 m), wave Reynolds number (7×10^3 - 2×10^6) and other surface ocean parameters. We also examine the size distribution and closure in aerosol number fluxes at different sea states.

2. Experimental

The Penlee Point Atmospheric Observatory (PPAO, http://www.westernchannelobservatory.org.uk/penlee/) on the southwest coastal of the United Kingdom has proven to be a suitable site for eddy covariance measurements of air-sea transfer (Yang et al. 2016a, 2016b, 2019). PPAO sits about 11 m above mean sea level and a few tens of m away from the water's edge. The eddy covariance (EC) system, including the fast aerosol sensors and a sonic anemometer (R3, Gill), is mounted on a mast at about 18 m above mean sea level. An ultra-fine condensation particle counter detecting aerosols > 1.5 nm radius (CPC-3025A, TSI) and a compact lightweight aerosol spectrometer probe (CLASP, Hill et al. 2008) providing size spectra for radii between ca. 0.1–6 μm at ambient humidity were employed at PPAO. The CPC was used for flux measurements between 24 February and 3 June 2015. The CLASP unit deployed between 17 February and 1 March 2015 had a size range of 0.11 to 6 μm. This encompasses a one-week of overlap with the CPC, which we primarily use to study the closure of aerosol number fluxes. A slightly different CLASP unit was deployed between 21 December 2016 and 16 February 2017, which had a size range of 0.15 to 6 μm. Because of the narrower size range of this second unit and thus more undetected film drop aerosols, data from the 2016-2017 period were only used to contrast between open water and fetch-limited conditions. In this paper, we operationally use the word "total" to refer to aerosol source flux (i.e. corrected for deposition, see below) either from the CPC or integrated over the entire CLASP size range, unless specified otherwise.

Previous EC observations of momentum, sensible heat, CO_2, and CH_4 fluxes at PPAO show that two wind sectors are representative of air-water transfer: the southwest sector for which airflow is from the open ocean, and the northeast sector with airflow from the fetch-limited Plymouth Sound (Yang et al. 2016a; 2016b; 2019). A flux footprint model for spatially homogeneous conditions (Kljun et al. 2004) predicts that under typical southwesterly winds, the majority of the turbulent flux at a sensor height of 18 m above mean sea level comes from waters several hundred meters upwind of the site with a mean water

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depth of ~20 m. When winds are from the northeast, the flux footprint is over the Plymouth Sound and does not overlap with

325   land on the opposite side of the Sound (5–6 km away) except possibly under strongly stable conditions.

In the eddy covariance method, aerosol number concentration (C) measured at high frequency (here 10 Hz) is correlated

with the vertical wind velocity component and averaged over time to yield the net aerosol flux (= $\overline{\rlap{\diagdown}{\diagup}}$, where primed quantities

are perturbations from the mean and the overbar is the averaging operator). The measured net flux is the sum of the source

(upwards, positive) and deposition (downwards, negative) flux components (Nilsson et al. 2001; Geever et al. 2005; Norris et al.

330   2013b). The sea spray source flux must be derived from the net flux by subtracting a deposition flux (= $\overline{C} \cdot V_d$). Here V_d,

negative in sign, is the aerosol deposition velocity from Slinn and Slinn (1980) (their Equation 4), which accounts for

gravitational settling and is computed assuming the density of sea salt. Both the CLASP and the CPC fluxes are initially

computed as 10-minute averages and then filtered for non-stationary turbulence conditions following Yang et al. (2016a). Valid

fluxes can be averaged (e.g. to hourly intervals) to reduce noise.

335   For the CLASP, a small fraction of the aerosols are lost to the short inlet (~25 cm) during the 0.1 s of transit time. The

loss is size dependent and predicted inlet efficiency varies from essentially unity for aerosols smaller than 2 μm radius (film

droplets and most of the jet droplets), to ~0.5 at 5 μm, and to ~0.1 at 8 μm (Pui et al. 1987). Spume aerosols are likely too large

to be efficiently sampled by the CLASP. We corrected for aerosol loss in the CLASP inlet prior to the flux calculations.

Following Norris et al. (2012), the CLASP measurements are converted from net fluxes to source fluxes using the size-resolved

340   V_d. Integrated over all CLASP sizes, this deposition correction amounts to -25 cm^{-2} s^{-1} in the mean (up to ~-200 cm^{-2} s^{-1}) when

winds were from the southwest. This represents 14% of the net flux on average (up to ~50%). For submicron aerosols only (i.e.

radius of 0.1–0.5 μm), the deposition correction is -7 cm^{-2} s^{-1} in the mean. Humidity flux induces a bias in the CLASP aerosol

flux (Fairall et al. 1984). This is because the particles are sized at ambient humidity, but grow/shrink with local relative humidity

(RH). We correct for this bias using the modified bulk correction scheme described by Sproson et al. (2013). The final CLASP

345   fluxes are presented at a constant relative humidity of 80% following the aerosol growth rate for sea salt reported by Gerber

(1985). Together these corrections amount to typically 20–30% of the total CLASP source number flux at PPAO. A robust

humidity observation was unavailable from December 2016 to February 2017. Thus the CLASP measurements during that phase

could not be fully corrected for humidity effects and were thus more uncertain.

The ultra-fine CPC sub-sampled from a Teflon tube (18 m long, 0.64 cm ID, flow rate of ~15 liters per minute). The

350   use of Teflon tube is generally not recommended for aerosol measurements, as its non-conductive nature can lead to significant

aerosol losses; it was employed here out of convenience because the CPC was sub-sampling from an existing inlet tube for CO_2

flux measurements (Yang et al. 2016b). A brief test of CPC measurements at PPAO between using the long Teflon inlet tube

and a ~5 m, 0.32 cm ID stainless steel tube did not show any obvious difference in the aerosol number concentration. The CPC

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cospectra also do not suggest severe aerosol losses at high frequencies. The better than expected transmission through the Teflon
sampling tube may be because the tube had been used under high flow rate at PPAO for nearly a year prior to the CPC
measurements. It was well coated with sea salt, which probably increased its conductivity and thus reduced electrostatic aerosol
losses.

The long sampling tube resulted in a delay time of ~3 s in the CPC signal relative to the turbulent wind measurements,
as determined by the maximum covariance method between aerosol concentration and vertical wind velocity. This delay is close
to the expected time based on inlet length, diameter, and flow rate in the main tube (2.3 s). The CPC inlet efficiency, accounting
for its length, a 90° turn and three bends (not including any electrostatic losses), is predicted to be essentially unity for 0.3 <
radius < 2 μm (Pui et al. 1987). For radius < 0.3 μm, the mean inlet efficiency is 0.96 (<0.8 for nucleation mode aerosols). For
radius > 3 μm, the efficiency drops to ~0.7. Previous observations in the marine atmosphere show that total aerosol number is
usually dominated by particles below a radius of about 0.3 μm (e.g. Hoppel et al. 1990), which suggests only a minor inlet loss
for the CPC (≤ 4%). We choose not to apply any inlet efficiency correction to the CPC flux since the corrections are probably
small and the size distribution of aerosol flux below 0.1 μm (lower cutoff of CLASP) is not known. We correct our CPC fluxes
for high frequency flux attenuation due to the finite instrument response time (1 s for 95% change) using an empirical filter
function approach analogous to that for gas flux measurements (e.g. Yang et al. 2013). Increasing with wind speed, this
correction amounts to ~10% of the CPC number flux for the average condition at PPAO.

The deposition correction for the CPC flux is only approximate due to both uncertainties in V_d and the lack of
knowledge of the fine/Aitken mode aerosol size distribution. The deposition velocity for submicron aerosols over water,
dependent on aerosol size and environmental conditions, is on the order of approximately 0.01 to 0.1 cm s^{-1} (Slinn and Slinn
1980; Duce et al. 1991). The aerosol size distribution below 100 nm radius was not measured at PPAO but previous maritime
observations of submicron aerosols generally suggest peak number concentrations at radii of ~25 and 100 nm (e.g. Hoppel et al.
1990). We compute the wind speed dependent V_d at these two aerosol sizes using the Slinn and Slinn (1980) parameterization,
which amounts to 0.034 and 0.010 cm s^{-1} for the mean conditions at PPAO. For simplicity, we take the average of the two V_d
datasets and multiply it by the CPC number concentration to estimate the deposition flux. When winds were from the southwest,
the deposition flux amounts to 33 cm^{-2} s^{-1} in the mean (19% of the net flux).

Frequency weighted cospectra of total CLASP and CPC net aerosol fluxes averaged to wind speed bins are shown in
Figures 1 and 2 for the open water wind sector. In low-to-moderate winds, the aerosol cospectra are broadly consistent with the
theoretical spectral shape for turbulent transfer (Kaimal et al. 1972) and with previously observed gas cospectra at PPAO (Yang
et al. 2016a). With increasing wind speeds the magnitudes of the cospectra increase, reflecting greater sea spray fluxes. The
CPC cospectra are much noisier than those of the CLASP. This may be because most of the aerosols detected by the CLASP

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arise from sea spray emission. In contrast, only a very small fraction of the aerosols detected by the CPC participate in rapid air-

385   sea exchange (i.e. sea spray emission); the vast majority comes from other sources (e.g. pollution) and increases the random

measurement noise in the CPC flux (see analogous discussion on methane flux by Yang et al. 2016a).

3. Results and Discussions

3.1 Sea Spray Flux Closure and Wind Speed Dependence

390   Aerosol fluxes were almost always positive (i.e. from sea to air), indicating sea spray emission. Figure 3 shows the time

series of total aerosol number concentrations and source fluxes from the CPC and the CLASP (during the one week of overlap),

relative humidity and the CLASP deposition correction term, significant wave height and tide height above datum, as well as

wind speed and wave Reynolds number. Winds were coming from the southwest for the majority of this week with peak speed >

16 m s^{-1}. The CPC detected baseline concentrations of a few hundred aerosols cm^{-3}, typical for a marine atmosphere. Many

395   short and sharp spikes (of the order of thousands cm^{-3}) are apparent in the CPC time series. These spikes are often coincident

with spikes in sulfur dioxide and carbon dioxide and are likely from ship exhaust emissions. CPC fluxes during such brief

periods of excessive variability (i.e. relative standard deviation over 50%) are removed on the basis of non-stationarity.

Aerosol number concentration integrated over all sizes is much lower and more constant from the CLASP. For the

southwest wind sector, the median supermicron and submicron aerosol concentrations detected by the CLASP were 11 and 24

400   cm^{-3} respectively. The magnitude of the former is consistent with the typical sea spray aerosol number concentration in the

marine boundary layer (e.g. O'Dowd and Smith 1993). The CLASP did not detect ship plumes as ship emitted particles tend to

be small (< 30 nm radius) and below the measurement size cutoff (e.g. Hobbs et al., 2000; Petzold et al., 2008). Despite the

large differences in total number concentrations between the CPC and CLASP, total aerosol fluxes from the two instruments

were generally comparable in magnitude. The CPC fluxes were slightly higher than the integrated CLASP number fluxes by a

405   mean (median) difference of 45 (40) cm^{-2} s^{-1} during this week. As discussed in Section 3.3, this difference is likely due to the

part of the film drop mode not completely captured by the CLASP as a result of its lower size cutoff (~0.1 µm).

Sea spray flux peaked during periods of high winds and large waves. At low tide, the distance between the water's edge

and the flux sensors increases, shifting the center of the flux footprint closer to the shoreline. Previously observed transfer rates

of momentum, sensible heat, and gases (CO$_2$ and CH$_4$) did not vary with the tidal height (Yang et al. 2016a, 2016b), suggesting

410   that the narrow surf zone (width of a few m) beyond the rocky shoreline in front of PPAO has a negligible influence on the

measured fluxes. The same appears to be true for the aerosols, as periods of low tide do not consistently result in large sea spray

fluxes (Figure 3). Thus unlike Geever et al. (2005), we do not need to filter out data for low tide conditions. On 27 February

2015, during light, northerly winds from over the land, aerosol fluxes from both instruments were near zero, as expected.

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Figure 4 shows the wind speed dependence in aerosol source flux from the CPC (February to June 2015) and from the CLASP (February to March 2015). Here we have restricted data to the southwest wind sector (open water) only. Across most of the wind speed range, total aerosol source fluxes from the CPC and the CLASP show a similar relationship with wind speed. Sea spray source flux from PPAO amounts to about 200 cm^{-2} s^{-1} in the mean (median of ~150 cm^{-2} s^{-1}), increasing non-linearly with wind speed up to ~1000 cm^{-2} s^{-1} at a wind speed of 20 m s^{-1}. These fluxes are in reasonable agreement with the source flux relationship found by Geever et al. (2005; >5 nm radius) for a coastal site at Mace Head during high tide, when wave breaking at the shore did not unduly influence their measurements. In comparison, the relationship found by Nilsson et al. (2001; >5 nm radius) from the Arctic Ocean is significantly higher than the PPAO measurements at wind speeds above ~12 m s^{-1}. Bin-averages of total aerosol number flux measurements at PPAO are plotted on a log scale against wind speed in Figure 5 along with the parameterizations from Nilsson et al. (2001) and Geever et al. (2005) for both net and source fluxes. Measured net fluxes at PPAO agree very well with the net fluxes from Geever et al. (2005). The source fluxes show greater discrepancy due to the different deposition correction schemes applied.

3.2 Non-Wind Speed Controlling Factors on Sea Spray Fluxes

Sea spray flux is more strongly associated with significant wave height (H_s, derived from the full wave spectrum) than with wind speed (Figure 6). Wave data when winds were from the southwest are taken from a Waverider buoy from Looe Bay, about 16 km west of PPAO (http://www.channelcoast.org/data_management/real_time_data/charts/?chart=98). The stronger dependence on H_s is reflected in the hourly CPC data (n = 452) by both higher R^2 (0.65) as well as the Spearman's rank correlation coefficient (R = 0.71) compared to the dependence on wind speed ($R^2 = 0.47$ and Spearman's R = 0.51). When H_s was below 0.5 m, the mean CPC source flux averaged to about zero. Such a strong dependence of sea spray flux on wave height is typically not observed over the open ocean (e.g. Norris et al. 2013b), and is likely due to coastal wave breaking. Equilibrium wind waves at wind speeds > 10 m s^{-1} as well as swell have wavelengths that are longer than twice the water depth within the PPAO flux footprint (Pierson and Moskowitz, 1964), and should shoal due to interactions with the sea floor. We thus expect coastal waves to break more frequently and generate more sea spray compared to open ocean waves at the same wind speeds.

Following Zhao and Toba (2001), we computed the wave Reynolds number as $R_{Hw} = u_* \, H_s / \nu$, where u_* is the friction velocity from eddy covariance and ν is the kinematic viscosity of seawater. Sea spray flux increases approximately linearly with the wave Reynolds number (R_{Hw}, Figure 7). The source flux dependence on R_{Hw} is slightly weaker than on H_s but stronger than on wind speed ($R^2 = 0.62$ and Spearman's R = 0.63) for the open water sector. Norris et al. (2013b) found a linear dependence between sea spray flux and R_{Hw}. They also argued that below a critical R_{Hw} of 7.2e^4, wave breaking does not occur and sea spray source flux should be zero, in general agreement with observations here. The linear dependence of sea spray flux at PPAO on

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R_{Hw} is qualitatively consistent with their open ocean results. The R_{Hw} parameterization from Norris et al. (2013b) integrated over

445　all CLASP size bins is shown in Figure 7. We see that sea spray fluxes measured at PPAO exceed those open ocean

observations by about an order of magnitude (but are smaller than estimates over a surf zone by Clarke et al., 2006, as shown in

Section 3.3).

　　　　The influence of waves on coastal sea spray generation is further illustrated in the comparison between open water and

fetch-limited conditions (Figure 8). We use data from the second CLASP deployment (December 2016 to February 2017) for

450　this analysis, as winds were seldom from the northeast during the first CLASP deployment (February-March 2015). Here we

separate the integrated sea spray fluxes measured by the CLASP into two different wind sectors: the southwest (open water) and

the northeast (facing the Plymouth Sound with a fetch over water of ~5 km). At a given wind speed, sea spray fluxes were

generally greater for the open water sector than for the fetch-limited sector. Wind speed was an even poorer predictor of sea

spray flux from the open water during this period, as some high sea spray fluxes were observed at low wind speeds due to the

455　presence of large swell. Fluxes from both wind sectors show better correlations against H_s, though with different trends; higher

fluxes are observed for the fetch-limited conditions than for open water at a given wave height. Here H_s in the Plymouth Sound

is predicted using a parameterization for fetch-limited waters (Resio et al. 2002) as a function of fetch and friction velocity since

a direct wave measurement was not available. The different functional dependencies of the sea spray flux on wind speed and

wave state for open water and fetch-limited conditions are reconciled when their joint influence is accounted for by the wave

460　Reynolds number. When plotting against R_{Hw} sea spray fluxes from both wind sectors fall closely on the same curve (Figure

8C). This clearly illustrates the importance of jointly accounting for the influence of both wind and waves on air-sea exchange.

This result is consistent with the recent findings of Brumer et al. (2017a,b) for whitecaps and gas transfer, and from Norris et al.

(2013b) for sea spray fluxes.

　　　　There is an observable sea-state dependence in the sea spray flux for the open water sector. Here we separate the CPC

465　flux data into two groups of sea states: increasing wind speed (i.e. hourly increase by more than 1 m s^{-1}) and decreasing wind

speed (i.e. hourly decrease by more than 1 m s^{-1}). These correspond approximately to younger/growing seas, and older/more

developed seas, respectively. As shown in Figure 9, at intermediate wind speeds (~10 m s^{-1}) the aerosol flux is in the mean about

twice as high during periods of decreasing wind speed than during periods of increasing wind speed, qualitatively consistent with

Norris et al. (2013b) for sea spray fluxes and Callaghan et al. (2008) for whitecap fraction. H_s is also larger during periods of

470　decreasing wind speed. Waves shoal and are should be likely to break near the coast regardless of their state of development.

Thus larger waves from more developed seas tend to lead to greater sea spray fluxes in these kinds of coastal environments.

　　　　It is interesting that there is a large discrepancy in magnitude in the source flux vs. R_{Hw} relationship between the open

ocean observations from Norris et al. (2013b) and those at PPAO (Figure 7) — if R_{Hw} is such a good predictor of air-sea fluxes,

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why does it fail to reconcile these two data sets? We suggest two possible reasons. First, shoaling may result in more frequent

475   and intense wave breaking near the coast compared to the open ocean as the waves steepen upon approaching the shore (e.g.

Elgar et al., 1997). Second is a potential difference in aerosol production per unit area whitecap between a coastal region with

shoaling waves and the open ocean as a result of the different wave breaking and bubble generation processes (Deane and

Stokes, 1999; Lewis and Schwartz, 2004; de Leeuw et al. 2011). Bubble populations near the sea surface were generally found

to be higher close to the coast than in open water (Johnson and Cooke, 1979; Brooks et al. 2009). Different void fractions have

480   also been observed beneath plunging and spilling breaking waves (Rojas and Loewen, 2010). Testing of these hypotheses

requires observations of the relationships between whitecaps, bubbles and R_{Hw} in the near-shore region.

Laboratory measurements suggest dependence of the sea spray flux on water temperature and salinity (e.g. Martensson

et al. 2003; Salter et al. 2014), while Tyree et al. (2007) showed that the addition of natural organic matter increased the

submicron aerosol flux by 50%. Previous observations of aerosol composition in marine environments imply that a significant

485   fraction of sea spray is made up of organic materials (e.g. O'Dowd et al. 2004). We investigate these dependencies by

comparing our sea spray flux data to surface ocean parameters from the marine station L4 (6 km south of PPAO) of the Western

Channel Observatory (http://westernchannelobservatory.org.uk/). From February to June 2015, sea surface temperature and

salinity varied between 9.2 and 12.5 °C and between 35.1 and 35.3, respectively. Chlorophyll a increased from 0.8 to 3.1 mg m^{-3}

during the spring phytoplankton bloom, while colored dissolved organic matter (from the E1 buoy 18 km offshore) also varied

490   by about a factor of four. We test the importance of these surface ocean parameters by examining their correlations vs. F_{SSA}',

where F_{SSA}' is the total sea spray source flux (F_{SSA}) minus a polynomial fit to F_{SSA} (as a function of wind speed or H_s). No

significant correlation was found. This suggests that within the range of conditions observed at PPAO during this deployment,

waves and wind are the first-order drivers for sea spray formation, while the other parameters appear to be of little importance.

3.3 Size Distributed Aerosol Number Concentrations and Fluxes

Figure 10 shows number distribution (dN/dR$_{80}$) vs. radius at a humidity of 80% (R$_{80}$) averaged in wind speed as well as

significant wave height bins. Data are taken from February–March 2015 and for the southwest (open water) wind sector only.

The overall distribution in concentration is fairly typical of the marine atmosphere, with most aerosols in the submicron mode.

The number distributions are more clearly segregated by significant wave height than by wind speed for radius up to about 2 μm.

500   Above this cutoff, the number distribution seems to be largely independent of H_s or wind speed.

In accordance with the total aerosol number fluxes, size distributed fluxes from the PPAO (dF/dR$_{80}$) are significantly

higher than measurements over the open ocean (Figure 11). For example, in moderate seas at PPAO the measured source flux at

R$_{80}$ of 1 μm exceeds 10^6 m^{-2} s^{-1} μm^{-1}, compared to approximately 10^5 m^{-2} s^{-1} μm^{-1} from Norris et al. (2013b). Sea spray flux

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from the coastal seas near PPAO is lower than estimates from the surf zone by Clarke et al. (2006). Readers interested in other previous measurements and parameterizations of size distributed sea spray source fluxes are referred to reviews by O'Dowd and de Leeuw (2007) and de Leeuw et al. (2011).

Size distributed aerosol number concentrations as well as fluxes peak at the lowest size bin of CLASP (0.1 μm radius at ~80% RH), near the typical mode center for film droplets (Martensson et al. 2003; Tyree et al. 2007). We can crudely estimate the contribution of film drop fluxes below the detection cutoff for the CLASP by fitting a log-normal distribution to the observed dF/dR_{80}. Here we assume that the observed dF/dR_{80} at a R_{80} of 0.1 μm represents the peak in the film drop mode. Integrating the log-normal fit from 1.5 nm to 0.1 μm yields the "missing" film drop flux (median value of ~40 cm^{-2} s^{-1}), which amounts to about 25% of the total measured CLASP number flux. This is consistent with the finding that the total aerosol number flux from the CLASP was lower than that from the CPC by a mean (median) of 45 (40) cm^{-2} s^{-1}. Future observations of the fine aerosol size distribution (e.g. by a scanning mobility particle sizer) at PPAO should provide more information on the robustness of our assumption above.

About 70% of the sea spray number fluxes measured by CLASP are submicron (at RH of 80%), with the vast majority of the aerosol number flux residing between radius 0.1 and 1.1 μm (Figure 11). Aerosols with radius greater than 2 μm make up only ~1% of the integrated CLASP number flux. The distribution of source number flux below a radius of about 2 μm is more clearly segregated by H_s than by wind speed, as with the aerosol number size distribution. Above 2 μm radius, size distributed fluxes no longer seem to depend on H_s or wind speed. There is a subtle decreasing trend in the ratio between film drop mode (radius of ~100 nm at 80% RH) and jet drop mode (radius of ~500 nm at 80% RH) with increasing significant wave height (Figure 12). An examination of normalized dF/dR_{80} (by the respective mean flux) shows that with greater wave height, the jet drop mode appears to increase more steeply than the film drop mode. A similar shift in the shape of size distributed aerosol flux with growing seas has been observed previously by Norris et al. (2013a). They showed that the bubble spectra change with wind speed, with the concentrations of small bubbles (which lead to jet mode aerosols) increasing more rapidly than the large bubbles (which lead to film drop mode aerosols).

4. Concluding Remarks

Eddy covariance measurements of sea spray fluxes originating from the shallow waters upwind of PPAO show that about 70% of the total detected number fluxes were submicron. A reasonable closure is found between the aerosol number flux from the CPC (>1.5 nm in radius) and the total number flux from the CLASP (0.1−6 μm in radius) after considering the incomplete detection of film mode aerosols by the latter. Sea spray fluxes from the open water wind sector at PPAO increase with wind speed with a dependence that is similar to previous coastal sea spray flux measurements at Mace Head (Geever et al.

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2005). Our observed fluxes are greater in magnitude than most previous open ocean measurements except those reported by

Nilsson et al. (2001), but are lower in magnitude than previous surf zone estimates (e.g. Clarke et al. 2006).

Sea spray formation in this coastal environment is strongly dependent on sea state. Both significant wave height (H_s)

and wave Reynolds number (R_{Hw}) are better predictors of sea spray fluxes than local wind speed. The importance of waves is

further confirmed by comparing sea spray fluxes measured from the open water sector to fluxes from the fetch-limited Plymouth

Sound, where waves were much smaller at a given wind speed. For both wind sectors, sea spray fluxes correlate with H_s more

strongly than with wind speed, but with two very different relationships. The wave Reynolds number (R_{Hw}) reconciles the fluxes

from the two sectors. This finding is consistent with those of Brumer et al. (2017a,b), who found R_{Hw} to be a much better

predictor than wind speed of both the whitecap fraction and gas transfer velocity in different wind/wave regimes.

Sea spray fluxes measured at PPAO (open water sector) are likely only representative of the nearest few km(s) from

shore. The median supermicron and submicron sea spray fluxes from the CLASP during February–March 2015 were about 60

and 100 cm^{-2} s^{-1}, respectively. The residence times of supermicron and submicron aerosols in a 500 m deep marine atmospheric

boundary layer (MABL) against dry deposition are of the order of 0.6 and 6 days (at a respective deposition velocity of -1 and -

0.1 cm s^{-1}, Slinn and Slinn, 1980). The residence time for submicron aerosols is likely further reduced by wet deposition (~3

days; Lewis and Schwartz, 2004). At these timescales, the expected steady state supermicron and submicron aerosol number

concentrations for a well-mixed MABL based on our measured fluxes would be on the order of 60 cm^{-3} and 300 cm^{-3},

respectively. These are significantly higher than the observed median supermicron and submicron aerosol concentrations from

the CLASP of 11 and 24 cm^{-3} for the open water wind sector. Clearly aerosol concentrations and fluxes are not in steady state in

this coastal environment, consistent with previous findings by Andreas et al. (2010) and Freire et al (2016). The fluxes are

enhanced near the coast due to increased wave breaking resulting from shoaling of waves in shallow water, while the

concentrations reflect both sea spray generated within the flux footprint as well as the aerosol sources and sinks further upwind.

To map out the spatial distributions of sea spray fluxes, measurement techniques such as eddy covariance from a ship and aerial

imaging of whitecap fraction (e.g. from an unmanned aerial vehicle) are needed.

Data availability

The data collection for this work was unfunded and thus the data will not be submitted to a central depository. Please contact the

corresponding author for the data.

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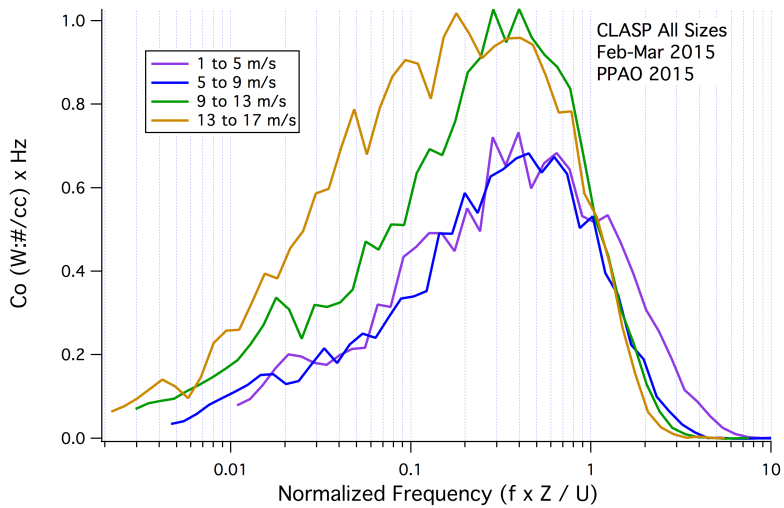
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Figure 1. Averaged CLASP frequency-weighted total (net) aerosol number cospectra in wind speed bins for the southwest (open water) wind direction.
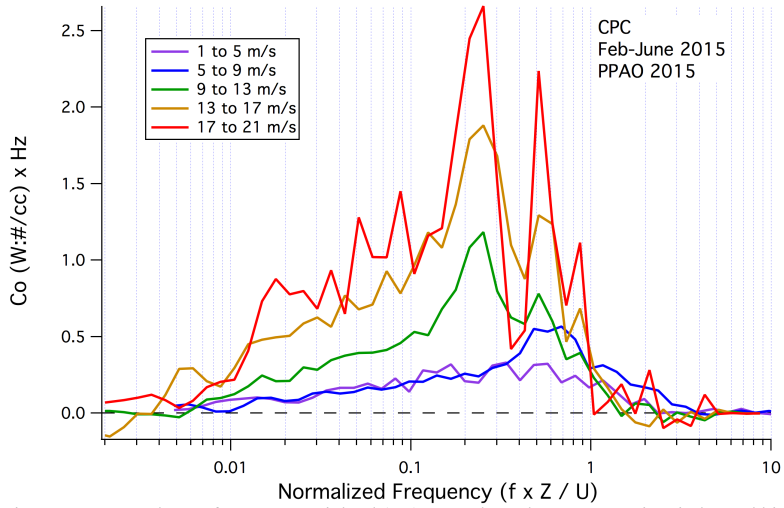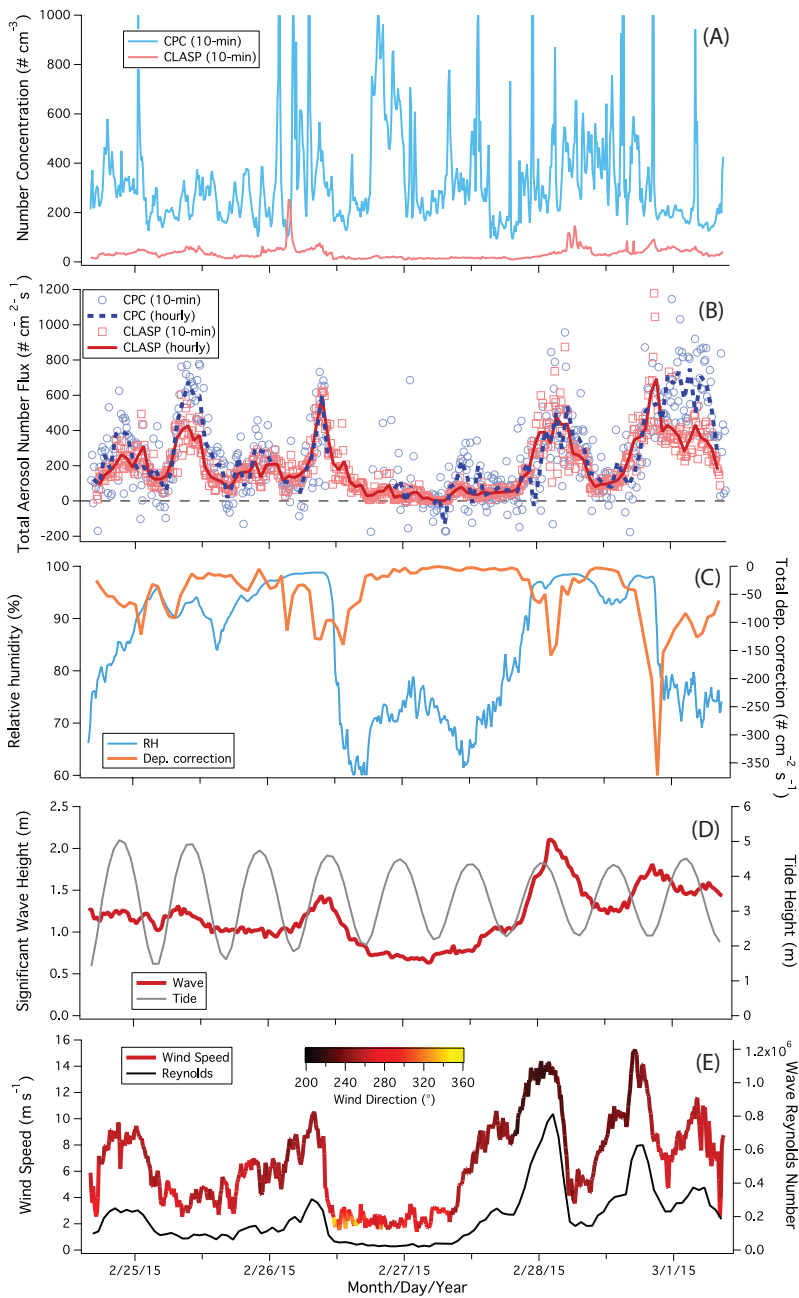

Figure 2. Averaged CPC frequency-weighted (net) aerosol number cospectra in wind speed bins for the southwest (open water) wind direction.

Figure 3. Times series of (A) CPC and integrated CLASP aerosol number concentration, (B) total aerosol number source flux from the CPC and CLASP, (C) relative humidity and the integrated CLASP deposition correction, (D) significant wave height and tide height, and (E) wind speed (color-coded by wind direction) and wave Reynolds Number.
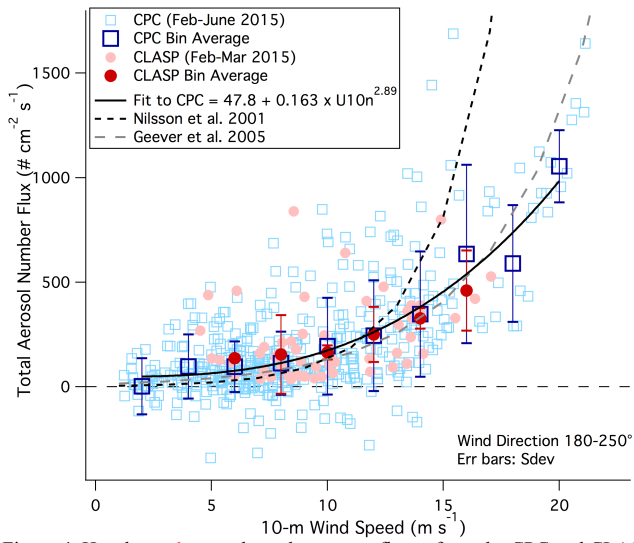
Figure 4. Hourly total aerosol number source fluxes from the CPC and CLASP vs. wind speed (data from 2015). Error bars correspond to standard deviations within the bins. Also shown are source flux relationships derived by Geever et al. (2005; > 5 nm radius) from the coastal site of Mace Head and Nilsson et al. (2001; > 5 radius nm) from the open ocean of the Arctic.
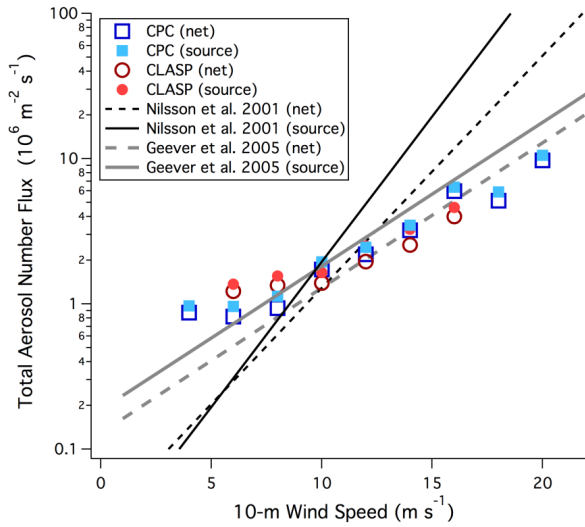

Figure 5. Bin-averaged total aerosol number fluxes (net and source) from the CPC and the CLASP (in log scale) vs. 10-m wind speed (data from 2015). Also shown are net and source flux relationships derived by Geever et al. (2005; > 5 nm radius) from the coastal site of Mace Head and Nilsson et al. (2001; > 5 radius nm) from the open ocean of the Arctic. Error bars (2 x standard error) are smaller than the marker size and thus not displayed.
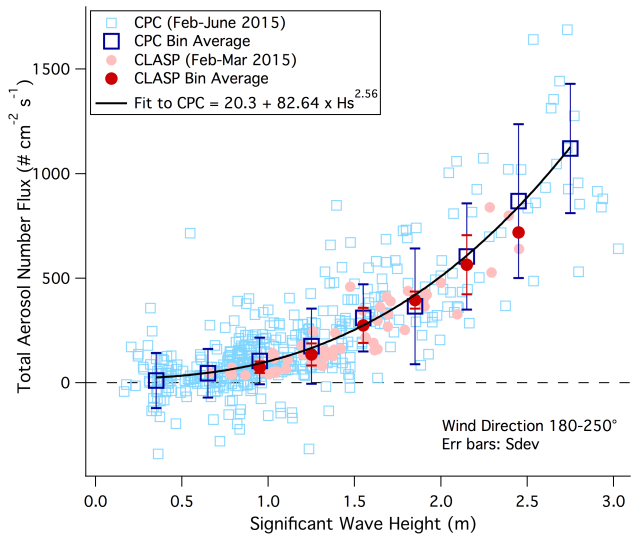
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Figure 6. Hourly total aerosol number source fluxes from the CPC and CLASP vs. significant wave height (data from 2015).
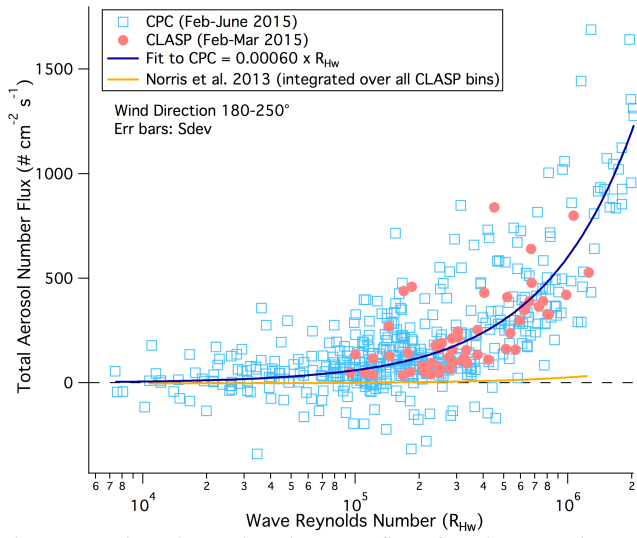
815   Figure 7. Hourly total aerosol number source fluxes from the CPC and CLASP vs. Wave Reynolds Number (log scale; data from 2015). Also shown are a linear fit to the CPC fluxes and the Reynolds Number parameterization from Norris et al. 2013 (integrated over all CLASP size bins).

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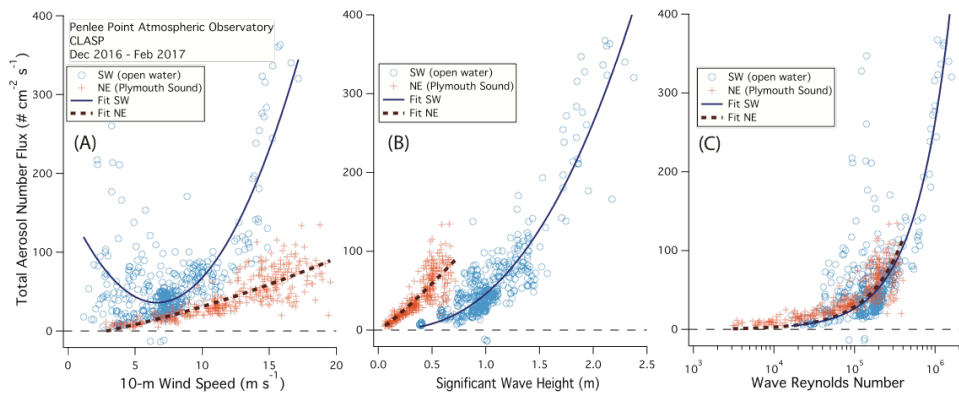
Figure 8. Total aerosol number <u>net</u> flux from the CLASP (December 2016–February 2017) vs. wind speed (A), significant wave height (B), and wave Reynolds number (C). Data are separated into two distinct wind sectors: the southwest sector that faces the open water, and the northeast sector that faces the Plymouth Sound (fetch of ~5 km). Power fits of aerosol fluxes to H_s and linear fits to R_{Hw} are also shown.
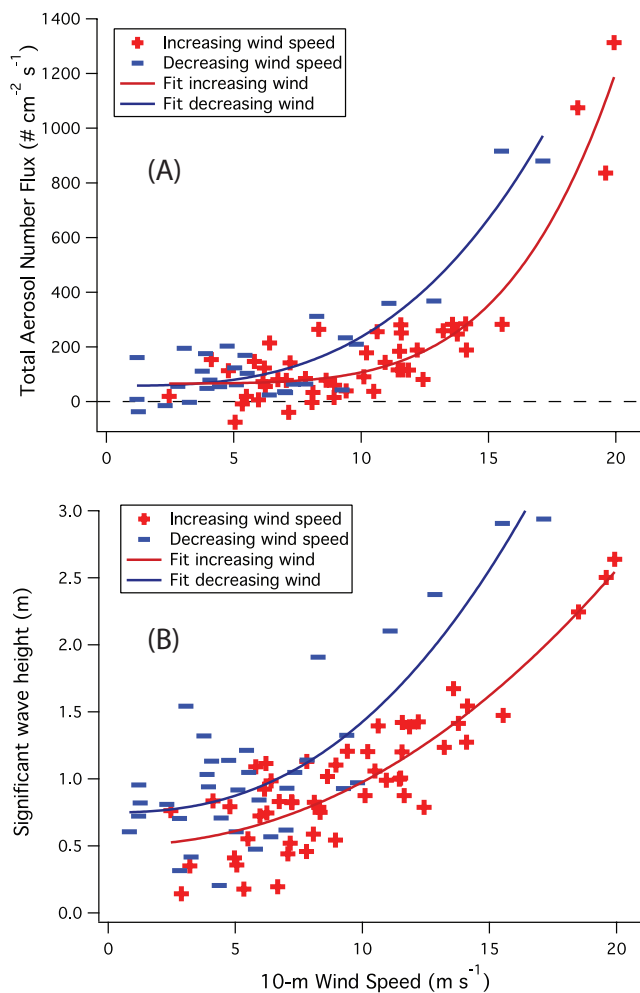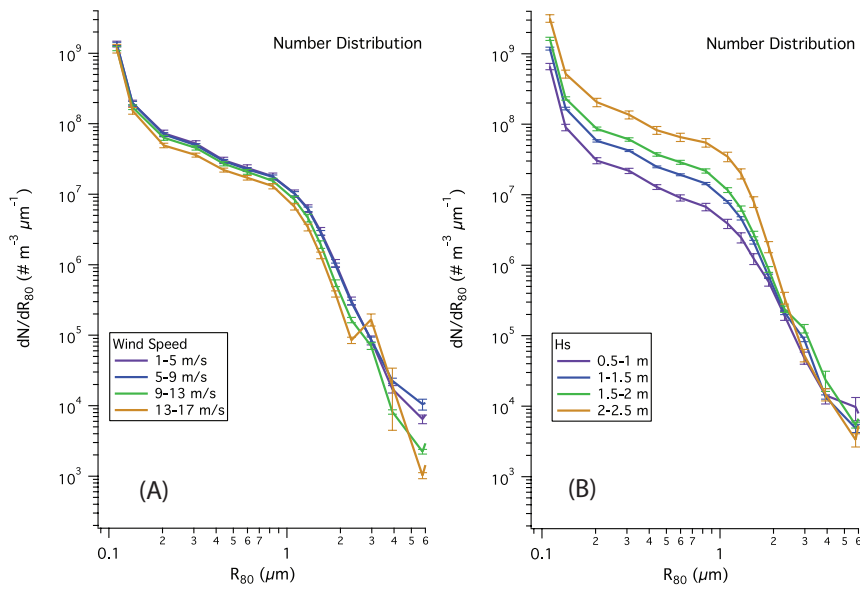
Figure 9. Total aerosol number source flux from the CPC for the southwest, open water wind sector versus wind speed (A) and versus significant wave height (B). Data on both plots are separated into periods of increasing wind speed (red crosses) and decreasing wind speed (blue dashes, see Section 3.2 for detail).
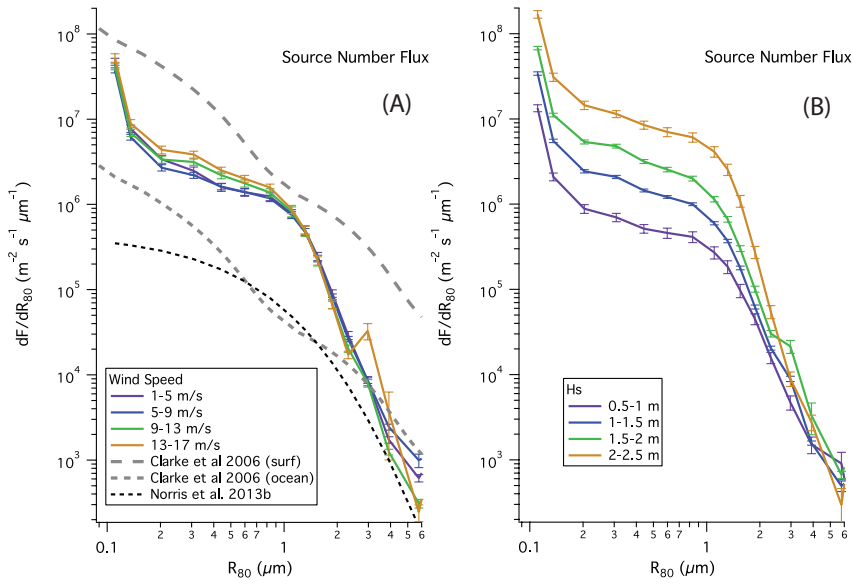
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Figure 10. Size distributed aerosol number from the CLASP, bin-averaged according to wind speed (A), and significant wave height (B). Data are limited to the southwest (open water) wind sector only. Error bars indicate standard errors. Radius adjusted from ambient humidity to a relative humidity of 80%.

Figure 11. Size distributed source number flux from the CLASP, bin-averaged according to wind speed (A), and significant wave height (B). Data are limited to the southwest (open water) wind sector only. The open ocean source flux parameterizations from Clarke et al. (2006) and Norris et al. (2013b) are approximated at a wind speed of 10 m s^{-1}, while the measured surf zone flux from Clarke et al. (2006) is more than an order of magnitude higher. Error bars indicate standard errors. Radius adjusted from ambient humidity to a relative humidity of 80%.
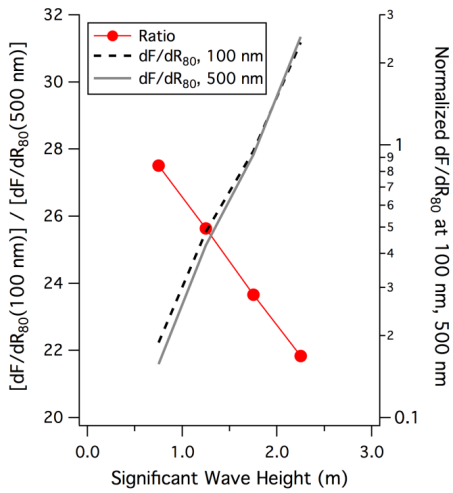
Figure 12. Ratio of size distributed source number flux in film drop mode (here R_{80} = 100 nm) to that in the jet drop mode (here R_{80} = 500 nm) as well as normalized dF/dR_{80} at these sizes as a function of significant wave height.