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Environmental Controls on Coastal Coarse Aerosols: Implications for Microbial Content and Deposition in the Near-Shore Environment

M. Elias Dueker,*[,†] Kathleen C. Weathers,[‡] Gregory D. O'Mullan,[†,§] Andrew R. Juhl,[†] and Maria Uriarte[∥]

[†]Lamont Doherty Earth Observatory, Columbia University, 61 Route 9W, Palisades, New York 10964, United States

[‡]Cary Institute of Ecosystem Studies, Box AB, Millbrook, New York 12545-0129, United States

[§]School of Earth and Environmental Sciences, Queens College, City University of New York, 65-30 Kissena Boulevard, Flushing, New York 11367, United States

[∥]Ecology, Evolution, and Environmental Biology Department, Columbia University, 2960 Broadway, New York, New York 10027-6902, United States

Supporting Information

ABSTRACT: Coarse aerosols (particle diameter (D_p) > 2 μm) produced in coastal surf zones carry chemical and microbial content to shore, forming a connection between oceanic, atmospheric, and terrestrial systems that is potentially relevant to coastal ecology and human health. In this context, the effects of tidal height, wind speed, and fog on coastal coarse aerosols and microbial content were quantified on the southern coast of Maine, USA. Aerosols at this site displayed clear marine influence and had high concentrations of ecologically relevant nutrients. Coarse aerosol concentrations significantly increased with tidal height (i.e., decreasing distance from waterline), onshore wind speed, and fog presence. As onshore wind speeds rose above 3 m s^{-1}, the mean half-deposition distance of coarse aerosols increased to an observed maximum of 47.6 ± 10.9 m from the water's edge at wind speeds from 5.5-8 m s^{-1}. Tidal height and fog presence did not significantly influence total microbial aerosol concentrations but did have a significant effect on culturable microbial aerosol fallout. At low wind speeds, culturable microbial aerosols falling out near-shore decreased by half at a distance of only 1.7 ± 0.4 m from the water's edge, indicating that these microbes may be associated with large coarse aerosols with rapid settling rates.

INTRODUCTION

The microbial component of atmospheric aerosols has long been recognized as a source for human allergies and disease transmission[1] and is thought to play an important role in the observed cosmopolitan distribution of some species of bacteria.[2] Important sources for microbial aerosol content include shedding from larger organisms;[3] long-range transport of dust particles;[4] and emissions from marine surface waters.[5] While the first two sources have been well studied, the ocean contribution of bacteria to the atmosphere has yet to be fully characterized, especially in the near-shore environment. A recent review estimated that 50−1000 bacterial cells m^{-2} s^{-1} are emitted from marine waters globally.[6] This may underestimate emissions from coastal waters, where microbial abundances can be higher than in the open ocean,[7] and aerosol production is increased by wave-shore interaction in the surf zone.[8] Further, it is in these coastal settings that onshore winds transport marine-derived nutrients and microbes to land, creating an interecosystem transfer that is rarely considered.[9]

Because of long atmospheric residence times (days to weeks) and long distance transport potential (100′s to 1000′s km), fine aerosols (particle diameter (D_p) < 2 μm) are the usual focus of atmospheric aerosol studies.[10] Bacterial aerosols, however, are most often associated with particles in the coarse aerosol fraction (D_p > 2 μm).[6] In coastal settings, coarse aerosol particles are primarily products of bursting bubbles in coastal surface waters created by wind-wave and wave-shore interactions.[5,8,11] Once formed, onshore winds transport these particles over land, where they are eventually deposited through gravitational settling, inhalation, or surface interception.

Previous studies have shown that particle size plays an important role in predicting chemical content[12] and microbial abundance and viability in atmospheric aerosols.[13] Despite this, the effects of physical factors including tidal movement, wind speed, and fog on the size distribution of coarse aerosols have rarely been studied in the coastal environment.[8,10,12,14] Even less is known about the impact of these physical factors on coastal microbial aerosols. Recent studies of coastal microbial aerosols do not simultaneously track coarse aerosol production and ambient aerosol size distributions,[15,16] making it difficult to evaluate environmental variability in production, transport, and deposition.

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This study examines the influence of tidal height (i.e., proximity to water's edge), wind speed, and fog presence on coarse aerosol particle size distributions, nutrient concentrations and microbial content across a coastal waterfront with the aim of characterizing this important connection mechanism between oceanic, atmospheric, and terrestrial systems.

METHODS

Study Site. This study was conducted on a south-facing rocky shore of Southport Island, ME, USA (N43.80261 W69.66841). Winds blow predominantly from the south, resulting in frequent advection fogs.[17] Data were collected during three field campaigns, June 28 - July 7, 2008, Sept. 8 - Sept. 14, 2008, and June 28 - August 4, 2009. Sampling of both physical and microbial factors were conducted at two fixed locations (25 m apart) within 9 m of the mean high tide line. As the tide moved out (46 m total at meteorological equipment station and 25 m total at the microbial station), a distance transect from the stationary samplers to the water's edge was created over time.

Tidal Height, Wind Speed, and Fog Presence. Tidal height data for the study site were time-corrected from Gulf of Maine Ocean Observing System (GoMOOS) buoy measurements near Portland, ME. One-minute wind speed, wind direction, humidity, temperature, and precipitation data were measured by a Vantage Pro2 Plus Weather Station (Davis Instruments, Hayward, CA) installed at the study site 5 m above ground level and within 25 m of all sampling. Fog presence/absence was determined using a combination of field observations and time-lapse photography.

Aerosol Particle Size Distributions. Aerosol particle concentrations were measured using a stationary Met One 9012 Ambient Aerosol Particulate Profiler (Met One Instruments, Grants Pass, OR) placed at a height of 2 m, within 9 m of the mean high waterline (55 m from the mean low waterline). One-minute aerosol particle size data were recorded in bins with boundaries of 0.3, 0.5, 0.7, 1, 2, 3, 5, and 10 μm diameter (D_p), with an estimated cutoff of D_p = 30 μm. This range of particle sizes covers both fine and coarse aerosol particle fractions.[10] The mean particle size per bin is used to reference each bin throughout this paper (e.g., D_p = 4 refers to the D_p = 3−5 μm bin). Particle sizes were not corrected for relative humidity (RH) due to the low RH variability (Table 1) but were separated between foggy and clear conditions.[18] Because particle concentration data were log-normally distributed, aerosol number and volume size distributions were calculated from the geometric means of particle concentrations pooled across field campaigns. Particle volume was calculated assuming spherical shapes, with radius $D_p/2$. All statistical analyses for this study were performed using R statistical software (R Development Project 2008).

Aerosol particle concentrations recorded at low tide, during calm (wind speed = 0) and fog-free conditions were used to create reference coarse aerosol concentrations, number and volume size distributions. The reference data were compared to subsets of the coarse aerosol data designed to isolate the influences of tidal height, fog presence, and onshore wind speeds (Supplemental Table 1). Briefly, data for the high tide subset were gathered at high tide under no fog, no wind conditions. Data for the fog subset were gathered during fog events with no wind and at low tide. Data for the wind subset were gathered at wind speeds of 5.5−8.0 m s^{-1} at low tide and no fog. Comparisons between these subsets and reference total coarse aerosol concentrations were performed using Welch's Two Sample *t* test of log-transformed data.

Table 1. Mean Meteorological Conditions during Field Sampling Events in Coastal Maine, USA

	6/28/08-7/7/08	9/8/08-9/13/08	6/28/09-8/5/09
minutes logged (n)	1056	1359	21603
temperature (°C)	17.51 ± 0.06	16.30 ± 0.03	17.87 ± 0.01
humidity (RH%)	94.48 ± 0.11	92.70 ± 0.10	95.01 ± 0.03
wind speed (m s^{-1})	0.90 ± 0.04	2.68 ± 0.04	0.66 ± 0.01
onshore winds (%)	59	78	60
fog events	7	3	18

At wind speeds >3 m s^{-1} the deposition of coarse aerosol particles was modeled using linear regression on log-transformed coarse aerosol concentration data. The half-deposition distance for coarse aerosols was calculated from regression parameter *a* (Supplemental Table 2) according to the formula

$$1/2\text{-}deposition\ distance = \ln(2)/\text{-}a \quad (1)$$

Aerosol and Ocean Surface Chemical Content. Fog droplets were captured for chemical analyses of coastal aerosols using duplicate passive fog collectors (PFCs).[19,20] The PFCs consisted of circular Teflon string (0.5 mm diameter) arrays mounted at 2 m on PVC frames. Fog droplets impacted on the Teflon strings were funneled into an opaque, UV-protected sterile collection bottle. Both PFCs were rinsed with sterile deionized (DI) water before and after each fog event. For chemical analysis, 10−50 mL of fogwater (depending on collected sample volume) were passed through a 0.22 μm Sterivex filter to remove bacterial cells and frozen until analysis. Ocean samples were gathered from the ocean surface (~0.5 m depth, 2−10 m from shore) on a daily basis and similarly processed. Aqueous chemistry analyses were performed by University of Maryland's Horn Point Analytical Service Lab.[21] To assess marine influence on the aerosol chemistry, all concentrations were normalized using sodium (Na^+) concentrations and then compared to ocean surface chemistry.[22]

Microbial Aerosols and Ocean Bacteria. Total microbial aerosols were sampled using duplicate SKC Bioaerosol Samplers (SKC, Eighty Four, PA) according to Fierer et al.[23] Total bacterial abundances in aerosols, fogwater, and ocean surface waters were determined by preserving 1−10 mL of bioaerosol sample, fogwater (captured in a sterilized PFC), or surface ocean (collected at <0.5 m depth) in 3% formaldehyde (final concentration). Samples were stored in the dark at 4 °C until cell counts were performed (within 8 months of collection) according to Noble et al.[24]

Culturability of microbial aerosols depositing near-shore was tested by exposing 2−4 replicate Petri dishes containing LB media (Luria Broth Agar, Miller, Fisher Scientific) to ambient aerosols for 30 min over a range of environmental conditions. Although not all bacteria are capable of growth under media-selected conditions, this method provided a relative measure of culturable microbial aerosol fallout. This method also served as an indicator of relative viability of the culturable microbial aerosols (e.g., damaged/dead culturable cells would not grow upon depositing). The exposures occurred at a station 10 m upwind from the particle profiler, 6 m from mean high waterline, and 25 m from mean low waterline. The plates were positioned as the highest

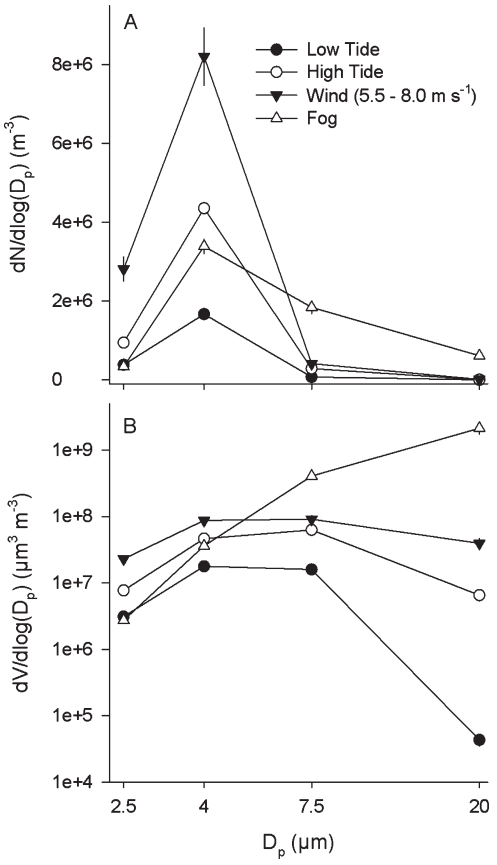

Figure 1. Comparison of observed coarse aerosol number (A) and volume (B) size distributions under differing environmental conditions (see text and Supplemental Table 1 for detail on data subsets), coastal Maine, USA. Note the log scale of the volume distribution *y*-axis and both *x*-axes. Vertical bars represent the standard error of the geometric mean of 1-min coarse aerosol concentrations (if not visible, the error interval is smaller than the plot point character). Because the standard error is calculated from a geometric mean, error bars may be uneven.

point on the shoreline (~1 m) to remove interception as a factor. After exposure, the plates were incubated at room temperature (17.7−20.3 °C). Colony forming units (CFU) were counted after 8 days. Microbial fallout (CFU m^{-2} s^{-1}) was calculated using the surface area of the exposed Petri dishes (0.0079 m^2) and the duration of exposure.

RESULTS

Drivers of Local Coarse Aerosol Concentrations and Size Distributions. During sampling this site experienced low mean wind speeds, high humidity, and frequent fog (Table 1). The number concentration of aerosol particles was dominated by fine aerosols, and the volume concentration was dominated by coarse aerosols (Figure 1, Supplemental Figure 1), as is normally the case for both marine and continental aerosols.[10] Coarse aerosol number size distributions were dominated by particles with D_p = 4 μm, and volume size distributions were dominated by particles with D_p = 4−7.5 μm, with the exception of foggy conditions (Figure 1). Relative to the reference condition, the number and volume of coarse aerosols increased during high tide. Moderate breezes (5.5−8.0 m s^{-1}) resulted in the greatest observed increases in both number and volume distributions for particles

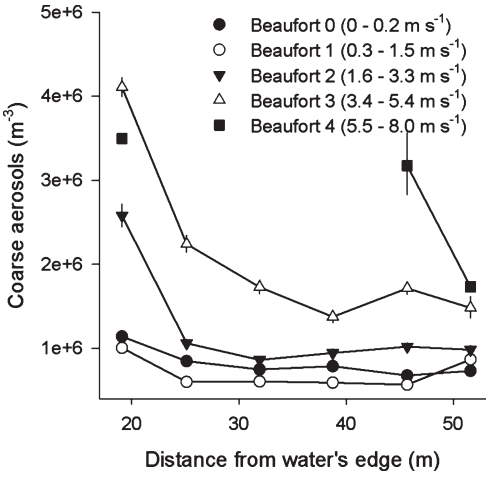

Figure 2. The geometric mean of 1-min coarse aerosol particle concentrations during fog-free conditions binned by distance from particle profiler to water's edge as mediated by tidal movement (high tide = 9 m, low tide = 55 m), coastal Maine, USA. Wind speeds binned according to Beaufort Scale categories. Vertical bars indicate standard error of geometric mean (if not visible, the standard error is smaller than the plot point character). Because the standard error is calculated from a geometric mean, error bars may be uneven.

with D_p = 2.5−4 μm. The presence of fog generated the greatest increases in both number and volume for coarse particles with D_p > 4 μm (Figure 1).

Tidal height (i.e., proximity to water's edge) had an important influence on the concentration of coarse aerosols. During periods of no wind or fog, the geometric mean of one-minute coarse aerosol concentrations at high tide (1.26 (±0.04) × 10^6 m^{-3}, n = 836 min) was significantly elevated (p < 0.01) over the low tide reference conditions (4.80 (±0.1) × 10^5 m^{-3}, n = 947 min). Fog coarse aerosol concentrations measured during times of no wind at both high and low tide were 1.94 (±0.07) × 10^6 m^{-3} (geometric mean, n = 1303 min), which was significantly higher than both low and high tide concentrations without fog (p < 0.01).

Onshore wind speed also influenced the concentration and size distribution of coarse aerosol particles (Figure 2). Wind speeds below 1.5 m s^{-1} gave low coarse aerosol concentrations that showed little decay, peaking at the closest and furthest distances from the waterfront (i.e., low and high tides). With wind speeds above 3.3 m s^{-1}, coarse aerosol particle concentrations were significantly elevated over the entire transect (Figure 2). At wind speeds above 3.0 m s^{-1}, the coarse aerosol half-deposition distance rose with increasing onshore wind speed, with an observed maximum of 47.6 ± 10.9 m from the water's edge at wind speeds from 5.5-8 ms^{-1} (Supplemental Table 2).

Aerosol Chemistry. The observed relationship between Na^+ (used as an ocean tracer) and chloride (Cl^-), bromine (Br^-), and magnesium (Mg^{2+}) concentrations in fogwater compared to known seawater stoichiometry[25] confirmed that coastal fogs incorporated sea salt aerosols (Table 2). Na^+-normalized Cl^- concentrations in fogwater, when compared to conservative seawater stoichiometry, reflected a 5% Cl^- loss. Fog water nutrient concentrations, including phosphorus, were high but within range of previously reported values (Table 2).[9,20]

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Table 2. Coastal Aerosol and Surface Ocean Chemistry, Maine, USA

	absolute concentrations				Na^+- corrected concentrations		
	coastal aerosols		surface ocean		coastal aerosols	surface ocean	aerosols/ocean
	n	mg L^{-1} (fogwater)	n	mg L^{-1}	(mg L^{-1})/(mg L^{-1})$_{Na+}$	(mg L^{-1})/(mg L^{-1})$_{Na+}$	
Cl^-	11	23.59 ± 4.41	20	16397.44 ± 73.80	1.70 ± 0.08	1.79	0.95
Br^-	10	0.11 ± 0.03	20	56.94 ± 0.26	0.006 ± 0.001	0.01	0.96
Mg^{2+}	18	3.24 ± 0.49	20	1087.66 ± 4.90	0.13 ± 0.01	0.12	1.09
	n	μM (fogwater)	n	μM	μM/(mg L^{-1})$_{Na+}$	μM/(mg L^{-1})$_{Na+}$	aerosols/ocean
DIN	22	282.40 ± 33.50	18	1.95 ± 0.30	18.20 ± 3.00	0.00020 ± 0.000030	91000
DIP	21	0.46 ± 0.11	20	0.45 ± 0.03	0.02 ± 0.01	0.00005 ± 0.000003	400
DON	10	37.85 ± 11.92	10	2.73 ± 0.55	1.00 ± 0.20	0.00030 ± 0.000200	3333
DOP	10	0.56 ± 0.10	10	0.20 ± 0.03	0.02 ± 0.004	0.00002 ± 0.000003	1000

Microbial Content of Surface Ocean, Coastal Aerosols, and Fog. Surface ocean bacterial concentrations averaged 9.2 (±1.0) × 10^5 cells ml^{-1} (n = 12). This finding agrees well with the global average reported for coastal waters of 5 × 10^5 cells ml^{-1}.[26] Fog bacterial concentrations averaged 9.7 (±6.3) × 10^4 cells ml^{-1} (n = 8), which is similar to the 1.0 × 10^5 cells ml^{-1} recently reported for cloudwater.[27] Total microbial aerosol counts were log-normally distributed and had a geometric mean of 1.6 (±0.8) × 10^6 m^{-3} air (n = 11). Tidal height, onshore wind speed, and fog did not have statistically significant effects on total microbial aerosol concentrations.

Microbial Fallout. Tide and fog significantly influenced the fallout of culturable microbial aerosols. Without fog, the mean microbial fallout was 0.06 ± 0.05 cfu m^{-2} s^{-1} (n = 29 plates, 4 events). Fog significantly increased microbial fallout (Welch's two sample t test, $p \ll 0.01$) with a mean of 1.3 ± 0.2 cfu m^{-2} s^{-1} (n = 69 plates, 5 events). The importance of fog in microbial fallout was also modulated by tidal height. Maximum microbial fallout was reached during foggy conditions at high tide, with a mean of 5.2 ± 0.8 cfu m^{-2} s^{-1} (n = 15 plates, 2 events).

Wind speeds during plate exposures averaged 0.9 ± 0.1 m s^{-1} and never exceeded 2.0 m s^{-1}. Under fog-free conditions, there was a significant positive relationship (linear regression on log-transformed data, n = 26, R^2 = 0.54, a = −0.41 ± 0.08, $p \ll 0.01$, b = 2.53 ± 1.00, $p < 0.05$) between proximity to the water's edge (tidal height) and microbial fallout (Figure 3). At these low wind speeds, microbial fallout decreased by 1/2 within 1.7 ± 0.4 m of the water's edge (calculated using regression parameter a in eq 1).

To demonstrate event-specific interactions between fog and coarse aerosol concentrations and microbial fallout, four plate exposure events are detailed in Figure 4. Coarse aerosol concentrations and microbial fallout were low in the absence of fog (Figure 4A, 4B). The initial onset of fog increased coarse aerosol concentrations by an order of magnitude (Figure 4B) and was coupled with increases by a factor of 3 or more in microbial fallout (Figure 4B, 4D). Wind speed, RH, and temperature during each exposure event had low variation and are plotted in Supplemental Figure 2.

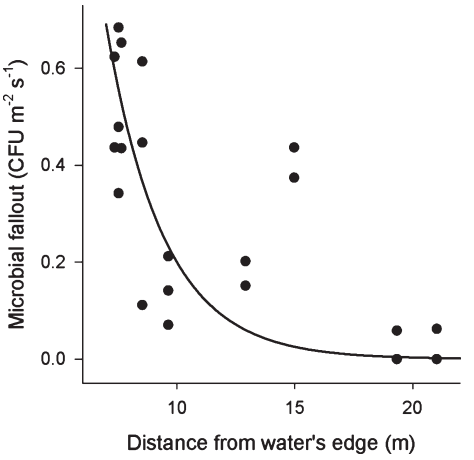

Figure 3. Fallout of culturable microbial aerosols under low-wind (<2.0 m s^{-1}), fog-free conditions plotted by distance of exposed plates to the water's edge as mediated by tidal movement (high tide = 6 m, low tide = 25 m), coastal Maine, USA. Each point represents a single plate count, and the decay curve is based on linear regression of log-transformed data.

DISCUSSION

Local Production and Transport of Coarse Aerosols from Coastal Ocean Surface to Land. Local ocean surface production of coastal coarse aerosol particles was confirmed by physical, chemical, and microbial characteristics of these particles. Coarse aerosol particle concentrations were notably increased at wind speeds >3.3 m s^{-1}, most likely due to local production from wind-wave interactions. These results agree well with increases in coastal coarse aerosol concentrations at wind speeds >4 m s^{-1} reported by Vignati et al.[12] De Leeuw et al. found that aerosol particles (D_p = 0.5-10 μm) were increased by 1−2 orders of magnitude in surf zones as the direct result of local aerosol production by breaking waves.[8] The negative relationship of coarse aerosol concentrations to distance from the waterfront across the range of wind speeds observed at this site (Figure 2) suggests that wind and waves locally produce coarse aerosols that are then transported over short distances, with over 1/2 of the particles settling out an observed maximum of 47.6 ± 10.9 m from the water's edge (Table 2). This pattern is likely to become more pronounced at higher wind speeds.

The aerosol particles captured in fogwater exhibited clear ocean influence, although the volumetric contribution of seawater to fogwater was small. The local nature of the sea salt aerosols captured by fog was demonstrated by the small loss of Cl^- when compared to Na^+ (5%) (Table 2). Once aerosolized from ocean water, up to 90% of Cl^- can be lost to atmospheric

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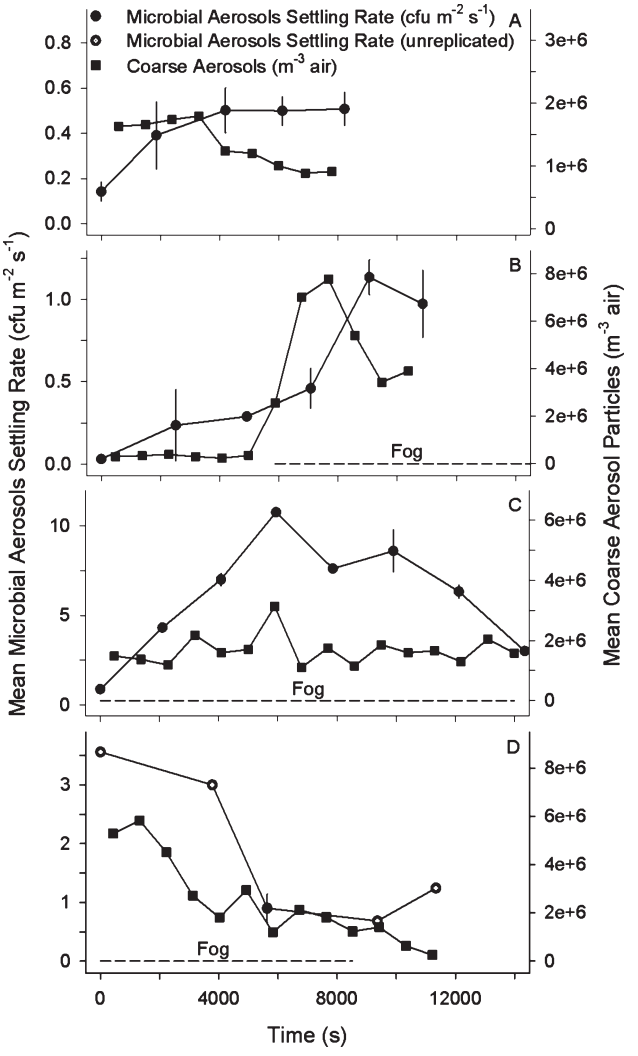

Figure 4. Event-based comparisons of total culturable microbial aerosols (left vertical axis) and 15-min coarse aerosol concentrations (right vertical axis), coastal Maine, USA. Please note differing vertical scale axes. The presence of fog is indicated by a horizontal dashed line above the horizontal axis. Vertical bars represent standard error of replicate plates (microbial aerosols) or 15-min coarse aerosol concentrations and if not visible are smaller than the plot point character.

chemical processes (particularly in reactions with HNO_3) over time in open ocean settings.[28] The much lower Cl^- loss reported here occurred despite high concentrations of dissolved inorganic nitrogen (DIN) in sampled fogwater chemistry (Table 2), indicating that sea salt aerosols measured in this study were recently produced, and therefore of local origin.

Finally, total microbial aerosols and microbial fallout also indicate local aerosol particle production. The average microbial aerosol loading of 1.6 ($\pm$0.8) $\times$ 10^6 cells m^{-3} was similar to that reported by Aller et al. sampling 4 m above a boat wake in Long Island Sound (2.0 $\times$ 10^6 cells m^{-3})[2] but over 2 orders of magnitude higher than that measured from an 85 m tower located 500 m from the coast of the UK (8 $\times$ 10^3 cells m^{-3}).[29] Also, microbial fallout at high tide (i.e., 6 m from the water's edge) was 3 times that at low tide (i.e., 25 m from water's edge) during low-wind clear conditions (Figure 3). The increase in microbial fallout with proximity to the water's edge suggests a local, marine source for these organisms and the aerosol particles with which they are associated.

Controls on Coastal Coarse Aerosols and Microbial Content. Tidal height, wind speed, and fog presence clearly affected the number and volume distributions of coarse aerosols at the waterfront (Figure 1). High onshore wind speeds created the greatest increases in aerosols with D_p = 2.5−4 μm. This range covers literature-reported median particle sizes (D_p = 2−4 μm) most often found to be associated with viable microbial aerosols in continental and coastal regions.[6] However, the low wind speeds (<2.0 m s^{-1}) encountered during agar plate exposures did not allow for adequate testing of wind effects on total microbial aerosol concentrations and culturable microbial fallout. Further research focusing on microbial aerosol fallout at the coastal interface for wind speeds >3 m s^{-1} is merited.

Proximity to the water's edge (as mediated by tidal movement) was found to significantly affect both coarse aerosol particle concentrations and microbial fallout. Both relationships closely approximated exponential decreases with distance from the waterline (Figures 2 and 3). Baylor et al. found similar trends when measuring the fallout of viral particles at a coastal site (half-deposition distance ~5 m) under windy conditions (although wind speeds were not specified).[30] Under the low wind conditions of this study, the small half-deposition distance for culturable microbial aerosols (1.7 m) compared to that found for coarse aerosols at the same wind speeds (77.0 m, Supplemental Table 2) strongly suggests that the microbes sampled were associated with large aerosol particles that settled out soon after production.

The local production of coastal coarse aerosol particles and culturable microbes measured in this paper underscores the importance of understanding the local deposition of these particles and microbes in the near-shore environment. Even under low-wind conditions, coarse aerosol particles containing both ecologically relevant nutrients and culturable microbes were transferred from ocean to land through tidal movement and surf-zone processes. Given evidence from coarse aerosol particle concentrations, this transfer should increase with importance when wind speeds exceed 3 m s^{-1}.

While previous studies have shown that fogwater contains culturable microbes and fungi,[31] this study is the first to report total bacterial loading of fogwater, particularly in a coastal setting. This is also the first report of increased fallout of culturable microbial aerosols during fog events. The increased fallout of culturable microbial aerosols may be both a function of increases in gravitational settling rates and increases in microbial aerosol viability. Environmental challenges to viability of aerosolized microbes include increased UV exposure, desiccation, temperature shock, radical pH reduction, and oligotrophic microconditions.[6] Fuzzi et al. postulated that fogwater could serve as a culture media for microbial aerosols, allowing them not only to survive the stressors normally associated with aerosolization processes but also to grow and metabolize while suspended.[31] While growth of microbes is not a valid explanation on the transport time scale of this study, increased viability from reduced environmental stress in fog water is likely.

Implications for Connecting Water Quality with Air Quality in Coastal Regions. We have established that even under low-wind conditions, there is a detectable movement of coarse and microbial aerosols from ocean to land, an important baseline understanding that will serve as the foundation for future quantification of the contribution of high winds to these fluxes.

These findings confirm the importance of coarse aerosols as a connection between oceanic, atmospheric, and terrestrial systems at coastal sites. Differences in bathymetry and coastal geometry may change the quantitative details of this process, but the general concept of measurable local coarse and microbial aerosol production and terrestrial deposition can be extrapolated to other coastlines.

The potential for aerosols originating from the open ocean to affect coastal urban air quality was recently investigated by Athanasapoulou et al.[32] Our findings suggest that aerosols created in coastal surf zones can also contribute to coastal air quality. This connection is relevant to the ecology of ecosystems and to human health in coastal regions, particularly in coastal urban centers, where dense populations are in close proximity to the waterfront, often in areas with poor water quality and high microbial loading in surface waters. Urban coastal waters often support bacterial abundances several orders of magnitude higher than coastal surface waters without anthropogenic influence. If these higher microbial concentrations lead to higher microbial aerosol concentrations (and particularly the aerosolization of sewage-related pathogens) this ocean-atmosphere-terrestrial microbial transfer point could have human health implications.

■ ASSOCIATED CONTENT

Supporting Information. Supplemental tables and figures referred to in the text. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author
*E-mail: med2109@columbia.edu.

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