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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simultaneous coastal measurements of ozone deposition fluxes and iodine-mediated particle emission fluxes with subsequent CCN formation" *by* J. D. Whitehead et al.

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We thank each of the reviewers for their encouraging comments and address each of their specific questions below.

Reviewer 1

1. vd for ozone – In the discussion about turbulent transport of ozone to surfaces, I was a bit surprised to see the statement: "There is little information on the processes governing rb above water and it is difficult to select an appropriate parameterization

for deposition to either the sea surface or the exposed sea floor." This part of the flux problem is the same for all gases. Hence all the literature regarding water vapor transport to water and land surfaces is relevant. So, I was expecting to see reference to classic papers like Kaimal et al., 1972, Kondo et al., 1975, etc.. Of course there are many complications for the open water case (fetch, wave state, etc.). The water vapor fluxes (in conjunction with a surface temperature measurement) would allow rb to be computed.. Anyway, this is a minor point, since as the authors state, rb is small compared to rs.

Kaimal, J. C., et al. (1972), Spectral characteristics of surface-layer turbulence, Q. J. R. Meteorol. Soc., 98, 563– 589.

Kondo, J. (1975), Air-sea bulk transfer coefficients in diabatic conditions, J. Boundary Layer Meteorol., 9, 91–112.

The first part of this sentence is perhaps a little misleading. What is meant is that a number of parameterizations for r_b exist in the literature all of which disagree with each other by some margin when applied to the measurements at Roscoff. As the reviewer states, open water is a complicated case. Therefore it was difficult to decide on a reliable parameterization for r_b, and to study this in depth would be time consuming and add nothing to the conclusions of the paper. This section will be clarified for the next version of the paper. However the paper by Kaimal et al. (1972) does not discuss the laminar sub-layer resistance, so we do not see how it can contribute to the discussion.

2. Figure 1 – the overlay obscured useful information in the photo. The overlay itself needs a plain background to be easily understandable.

This figure will be made clearer in the next manuscript.

3. "In order to ensure that the presence of the jetty wall was not influencing the air flow at the sensor location, the vertical wind angle was examined" – does this mean the *mean vertical wind was zero? Does "was not found to deviate" mean "did not deviate from vertical"? This could be written more clearly.*

The wind angle did not deviate by more than a few degrees from the horizontal. This will be clarified in the text.

4. With the caveat that I have zero expertise in this area. . . I am inherently dubious about particle flux measurements. If the particles are hydroscopic, they should be changing sizes in response to the local water vapor level on very fast (millisecond) time scales. That makes for a very complex signal to deconvolute, as there is a probably a large water vapor flux from the surface and the detectors respond to different sizes with different efficiencies. So, can one really obtain a unique solution to the flux? I realize this is a big subject and would rapidly consume a paper like this one. However, I think the reader needs some assessment of the uncertainty derived from this (and perhaps other) issues. Somewhere, the authors need to state their best estimate of uncertainty on the measured fluxes (for particles and ozone). This said, I think the paper's conclusions will hold up because the differences in particle fluxes between low flux and high flux conditions are orders of magnitudes.

Even if the particles were highly hygroscopic, the fluctuations in particle size due to fluctuations in RH on timescales of 0.1 seconds would be very small, and the effect on particle flux measurements would therefore be negligible. We will briefly explain this in the next manuscript.

5. p 20572, l21. "least empirical" is gratuitous

This will be changed to "simplest, most direct".

6. The wind angle criteria seems a bit generous. Allowing the sector to be along the coast means that given typical variance in wind directions, a large fraction of the

winds might be out of sector. Safer to put the cut-off at 60 degrees from the coast or something like that.

By contrast, the 60 degree cut-off is rather too stringent and would remove a large amount of our data and destroy the significance of our findings. In deciding on the bounds of the seaward wind sector, we took into account the shape of the coastline (which is not straight and veers away from the defined wind sector in both directions), and do not believe that the adjacent land holds much influence on the results. We will state this in the next manuscript.

7. 20575 "Various issues relating to sources of uncertainty, data quality control and in particular analysis techniques using the GFAS and similar instruments for ozone flux measurements, are discussed extensively by Muller et al. (2009)." Very awkward sentence, ambiguous.

We will change this sentence to read: "Use of the GFAS for ozone flux measurements, including data quality control, analysis techniques and sources of uncertainty, are discussed extensively by Muller et al. (2009)."

8. How do we know that the difference between high and low tide fluxes was not somehow due to the different height of the sensors? Are there control experiments where the sensor/intake height is varied within one tidal cycle or varied inversely with tidal height?

We agree with the reviewer that this is a complication. While no control measurements were conducted here, this will be a focus of further work. However the patterns in ozone deposition velocities and particle emission fluxes are more consistent with the picture of macroalgal emissions than with the effects of sensor height. Apparent particle emission fluxes, for example, are only seen during daytime low tide and are not seen during night time low tide. In addition, low tide apparent ozone deposition velocities were stronger during the daytime compared to night time due to photochemical destruction while there was no significant difference at high tide between daytime and night time. A paragraph will be added to the manuscript discussing this issue.

9. 20579. I would think that mixing induced by bottom shear (given the shallow depth of the water) would also enhance surface turbulence in this environment relative to that in the open ocean. Also line 14 remove parentheses

At high tide, the water was too deep (several metres) for bottom shear to be a significant factor. We will remove the parentheses.

10. Section 4.2 line17. The statement that direct deposition is "dominant" doesn't seem to make sense. Low tide resistance is about double that of high tide. So, direct deposition roughly equal to the other mechanisms, right?

The statement refers to night-time low tide conditions. At night time, in the absence of photochemistry, direct deposition is dominant in all tide conditions. Low tide (night time) resistance is half that of high tide (not double), because direct deposition to the sea floor (and constituent macroalgae) is faster than to the sea surface, as discussed clearly in the text.

11. "This may therefore be considered a lower limit to surface resistance to an exposed seafloor with non-uniformly distributed Laminaria beds amongst other species of seaweed." It is unclear exactly what "this" refers to. Anyway, it is not clear why it should constitute a lower limit, rather than an upper limit.

We refer to the surface resistance calculated by Kupper et al. (2008) for ozone deposition to the seaweed surface, and we agree that the text is not as clear as it should be. Their value constitutes a lower limit because it assumes uniform Laminaria seaweed surface to which ozone deposits, while in a coastal environment, there will be a more

patchy distribution, which will include bare seafloor and other species of seaweed. As deposition will be slower to non-seaweed surfaces, the calculated resistance over the heterogeneous seafloor will be higher. This will be clarified in the next version of the manuscript.

12. 20580 line 1 Should emphasize "gas phase photochemical destruction"

This will be added to the next version of the manuscript.

13. 20580 line 8 "fully consistent" is redundant. Either it is consistent or not.

The "fully" will be dropped in the next version of the manuscript.

Reviewer 2

1. The inset in Figure 1 does not seem to be referred to in the text and seems buried in the figure. Perhaps a stand alone figure that is referred to in the second paragraph of the introduction.

This figure will be made clearer in the next manuscript.

2. There is mention of typical corrections applied to the eddy correlation flux measurements but there is no mention of the magnitude of the corrections. The authors should also state their estimated final uncertainties for the ozone and aerosol flux measurements.

Corrections for routine corrections to fluxes, including time lag and coordinate rotations, are small and are of the order 1%. The Webb correction to the ozone fluxes resulted in an average increase in flux of 14%. Apart from the Webb correction, the other corrections combined were smaller than the estimated errors given in the paper for the averaged flux values associated with high tide, low tide, etc., and these will therefore represent the final uncertainties. The magnitude of the corrections will be briefly discussed in the text.

3. 20576; line 18; What percentage of the data was rejected?

Of the data collected during sea fetch conditions, around 66% were rejected due to non-stationarities and other quality controls. This will be explained in the text of the next manuscript.

4. 20569; line 28: "found" should be "showed"

5. 20574; line 22: remove the "were"

6. 20575;line 2: remove the +- symbol

7. 20577; line 2: remove "number of data points" n= should suffice

8. The discussion of figure 4 starts with figure 4b and ends with figure 4a. The figures should probably be the other way around.

9. 20578; line 23: should be "ranged from 0-7.8"

10. 20579; lines 12 and 14: insert a "the" before "open ocean"

11. 20579; line 25: either "during chamber measurements" or "in chamber measurement results"

12. 20580; line 18: rather "on a timescale"

These will be changed as requested in the next version of the manuscript.

Reviewer 3

1. While it is stated that the instrument boom was capable of being traversed vertically to accommodate the very large changes in tidal height, the resulting sustained

measurement height does not appear to be stated.

True. During the relevant flux measurement period, the sonic was at a fixed height of 3.4 m above the seafloor (corresponding to a tide height, when the water is at this level, of 5.6 m). This will be explained in the text of the next manuscript.

2. Figure 1. The figure should be edited to ensure the overlay is more clearly discernable.

This figure will be made clearer in the next manuscript.

3. The flux averaging period was 15 minutes. Some other works use a 30 minute averaging period. Can the authors comment on the choice of averaging period and any possible effects of this choice?

We checked that the flux averaging time was sufficient using the ogive method described in Foken (2006). This showed that 15 minutes was sufficient for capturing most of the flux. Too short an averaging time will result in failing to measure the low frequency contribution to the fluxes. Too long, and the steady state requirement of the eddy covariance technique may not be met, resulting in more instationarities. A brief discussion of this issue will be added to the manuscript.

4. 20574. Line 22: remove "were"

5. 20578. Line 23: change to "ranged from 0-7.8"

These will be changed as requested in the next version of the manuscript.

6. There is a significant difference between the two particle flux correlations, but the difference in sites and measurement setup could explain this. The referee agrees that further long-term investigations are required in this area (not necessary for this *publication).*

We thank the reviewer, and agree that the differences in site and measurement setup are factors here. We already describe the difference between the measurement sites in the discussion paper in terms of distribution of macroalgal sources, and stress the need for further investigation, which the reviewer agrees with.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20567, 2009.