O'Dowd and Ceburnis make some strong statements about our work. Some are due to misreading the paper (which means we weren't clear enough), some reflect real weaknesses in our paper, and some are simply incorrect. We'll start with the misunderstanding, since that seems to be the core of their objection. They state:

> The manuscript neglects many important works which provide very strong evidence pointing to a natural source of the discussed marine organics.

There is no doubt that the ocean can be a major source of organic matter in the atmosphere. As O'Dowd and Ceburnis point out, that has been demonstrated convincingly in many papers. What we found is that in air over the remote areas of the Pacific we covered, the amount of OM that could be clearly attributed to marine sources was surprisingly small. There is no reason to expect that to be true everywhere and we make no claims about that. It is likely that over large areas of the ocean—relatively unproductive central gyre regions—marine-sourced organics are a relatively small fraction of the aerosol. Because we were concentrating on tropical regions, we did give short shrift to literature from nearer the poles. An extended critical literature review is appended here and will be included in a much abbreviated form in the manuscript.

> The authors should demonstrate that, in the first instance, the AMS is operated correctly: were particles dried or sampled wet? It is normal to operate dry. If so, it is likely that organics internally mixed with sea spray will not be sampled due to sea salt bounce.

The methods section has been expanded a bit to answer questions about operation.

Did we miss organics that were actually present? Possibly, but not very likely. First, a very recent publication [Middlebrook et al., 2011] shows that our assumption of 50% collection efficiency was a bit optimistic: they recommend 45%. That would improve the agreement between size distributions and AMS total mass (Figure 2) from 0.81 to 0.90. That is well within the error of the measurements and suggests that we are not missing a very large fraction of the volatile mass.

Some organics, such as polystyrene latex, bounce around like the rubber balls they are and are thus poorly sampled by the AMS. While still a bit controversial, Hawkins et al. [2010] showed evidence that OM attached to dust particles can be lost , perhaps due to bounce, in the AMS. Perhaps the same is true with organics on NaCl crystals. On the other hand, both laboratory and ambient studies [Matthew et al., 2008; Middlebrook et al., 2011] show that liquid organics are collected with near unity efficiency and solids are collected with about 45±23(2σ)% efficiency, with most of the outliers high even when organics are present on crystalline ammonium sulfate. Those were from continental aerosol, so there is a small possibility that marine organics behave differently.

It is conceivable that primary marine organics are so refractory that the AMS fails to evaporate them at 600° C. Such stable OM certainly exists, but given the apparent carbohydrate-like composition found by Russell et al. [2010], primary marine organics are unlikely to be that refractory. After all, the pyrolysis products of the carboydrate cellulose are precisely what we see in the AMS when sampling biomass burning plumes. Indeed, Russell et al. [2010] found a noisy but nearly 1:1 ratio between FTIR and AMS OM in the far north Atlantic (their figure S3).

> You should be able to provide the mass spectra to support your claim on organics origin and demonstrate what organics are natural and anthropogenic. You should compare with Ovadnevaite et al. (2011) mass spectra. Why is the mass spectra not shown, isn't this the point of such an instrument?

We would have loved to show mass spectra of the organic component of clean marine aerosol! Unfortunately, our data suggest that we never found much, so we're not sure what we can claim our spectra represent. Some mixture of marine organics with poorly identified continental sources? Therefore we did not show any. We're happy to include some spectra in a supplement. Perhaps the possible biomass burning episode during the cruise would be of interest. Of course the spectra are completely dominated by sulfate fragments.

> The second reviewer also raises these critical points and additional points relating to the sensitivity of the SP2 – we are also surprised to see such high precision data presented without even a mention of the instrument in the instrumentation section.

The SP2 description has been expanded as requested. However, unless the SP2 was somehow giving false positive signals there is no way the issues raised by the second reviewer could affect the logic of this paper. Missing some of the BC mass would simply mean there was even more pollution present. False positives of consequence are very unlikely. They did not occur during periods with filters upstream of the SP2. Dust could in theory absorb enough to incandesce and Hawkins et al., [2010] did detect dust, but that would be an indicator of non-marine aerosol. An interesting conjecture might be that soot could accumulate on the ocean surface and be aerosolized as bubbles break, but we've never heard evidence of that (and the remote Southern and Equatorial Pacific would probably be less affected than most other regions).

An extended literature review, or evidence that small organic contributions to marine aerosol are entirely consistent with previous work.

Since much of the work on marine OM has been at higher latitudes, we gave that literature less emphasis than we should. It turns out that a more careful examination of the data presented in those papers is almost entirely consistent with our findings.

An issue here is that there is no such thing as pristine air. BC is a product of incomplete combustion and as such is an unambiguous indicator of non-marine aerosol. Since BC is concentrated in the accumulation mode, it is unlikely to have a much different lifespan than other combustion-related species. Therefore, whenever BC is found in a marine air mass, it must be accompanied by other aerosol products of combustion. Some fraction of the organics, nitrates, and sulfates must also be the product of combustion. The question is how much. This is true regardless of any definition of "clean" or "background" air. There are 3 obvious ways to handle this: try to subtract out the non-marine aerosol; develop a tracer that directly indicates the marine contribution; or ignore it, (making the implicit assumption that it is negligible).

Our original intent was to use the last option, figuring that we could find air that had been over the ocean long enough that much of the continental aerosol would be scavenged and marine OM would dominate. However, when we tried that, we found that the cleanest air (as determined from CO and BC) exhibited a striking relationship between OM and BC with an intercept near 0. That strongly suggests that the contribution of OM from marine sources was small.

Subtracting out an anthropogenic signal isn't necessarily straightforward. We saw an OM:BC ratio on the "clean" MBL of about 13:1. We don't have any reason to believe that is universal, as it must depend on sources and any scavenging that occurs. Reported OM:BC ratios near source regions range widely: 20:1 and 34:1 for southwesterly and northwesterly flow from the northeast coast of the US [Bates et al., 2005], 85:1 for fresh and 25:1 for aged Canadian forest fires [Singh et al., 2010], 1:1 from SW India and 8:1 from NE Asia [Quinn and Bates, 2005; INDOEX and ACE-Asia:polluted] , and 8:1 from southern Africa [Haywood et al., 2003]. Reid et al., [2005] show OC:BC ranging from 2 to 17 for a wide range of biomass burning, though they cluster around 11 or so.

There are a few papers that use the second option, using a marine tracer. Narukawa et al.,[2008] and Miazaki et al. [2010] use $\partial^{13}C$ to differentiate between marine and continental organics. Most of their samples are high-latitude and thus hard to compare to our findings, but the latter paper includes a few samples with back trajectories that originate over the oligotrophic central North Pacific before crossing narrow high-productivity areas to reach the ship they were using. It's not clear how much of the aerosol was contributed by the low-productivity region. They found that only 8-36% of the organic matter was due to marine sources. EC averaged 43 ng m^{-3}.

Ceburnis et al. [2011] take a similar approach, adding $\partial^{14}C$ to the analysis and using different values for continental $\partial^{13}C$. Yes it's convincing that they found marine organics, but we find very little about the origins of the air masses except that back trajectories (taken how often during the samples?) did not contact land for the previous 4-5 days. BC was clean by their standards, at 50 ng m^{-3}, but no attempt to correlate BC with non-marine OM was described. (It is also not clear what precautions were taken against sampling artifacts. It has long been known that quartz filter sampling is subject to both positive and negative artifacts due to vapors adsorbing to the quartz and semi-volatile aerosol species evaporating [e.g. Eatough et al., 1993; Turpin et al., 1994]. Perhaps enough aerosol was deposited to overwhelm vapor adsorption? It has been argued that temperatures at Mace Head are low enough that evaporation is unlikely. Is that the case for these samples? Filter blanks were mentioned, but not described; were they simply mounted and removed or was filtered air pulled through them for some period of time?)

Russell et al. [2010] found carbohydrate-like organic aerosol correlated with Na^+ from the far north Atlantic and Arctic Oceans. They found particles containing both Na^+ and OM. While not utterly conclusive, since cloud processing can join particles together, and wind speeds can be correlated with synoptic systems that bring particles from elsewhere, this constitutes pretty good evidence of marine organics. They had no BC measurements with the sensitivity of an SP2, but "the absence of long-lived biogenic and biomass burning VOCs ruled out most continental sources". Interestingly, the carbohydrate-like material they found resembles the carbohydrate-derived material found in biomass burning plumes. They even identified AMS fragments at m/z=60 and 44 as characteristic of primary marine aerosol--exactly the fragments common in biomass burning studies [Lee et al., 2010].

Perhaps the most conclusive demonstration of marine-derived organics is from Orellana et al., [2011], who developed an antibody probe specific to Arctic Ocean surface water material from the diatom *Melosira arctica* and demonstrated that its target was present in cloud water and aerosols. While a *tour-de-force*, it is not obvious that tropical oceans, with utterly different assemblages of phytoplankton species, would yield similar results.

However, most of the papers listed in the O'Dowd and Ceburnis comment take the last approach, declaring that conditions are "clean" and ignoring any possible OM associated with the 40, 50 or even 75 ng m^{-3} of BC. That is the case in Cavalli et al. [2004], the O'Dowd et al. [2004] paper based on it, Yoon et al. [2007], and O'Dowd et al. [2008]. Cavalli et al. [2004] show average OC:BC ≈ 43, comfortably above most source region ratios, so perhaps marine-origin organics dominate, but few of their back trajectories are from the central gyre of the North Atlantic, so there is no evidence that any of the organics derive from there. Yoon et al. [2007] chose 40 ng m^{-3} BC as limit of clean marine air, but did not report actual concentrations. Average total carbon was 0.4 μg m^{-3}, so average OC:BC may have been as low as 10:1. A significant fraction of that OC was likely due to the same combustion processes that produced the BC. It might be possible to go through the dataset and find substantial OC unconnected with BC from the central Atlantic, but this paper presents no proof. O'Dowd et al. [2008] built on Yoon et al. [2007], and did show a correlation between OC and a marine quantity [satellite-derived chlorophyll-a]. That relationship is compelling, but since no effort was made to exclude any OC associated with BC, the applicability of their best-fit line, and particularly the y-intercept, is open to question.

Sciare et al. [2009] certainly provide compelling evidence for marine-derived organic material at Amsterdam Island in the Austral summer, but during the winter they note a minimum in OM and a maximum in BC, which they credit to biomass burning in Madagascar and southern Africa. They do not speculate whether any OM accompanies that BC. In October/November, their BC averaged 6 or 7 ng m^{-3} and OC was roughly 100 ng m^{-3}.

Dall'Osto et al. [2010] is a particularly interesting paper to compare our results to, as they had a similar AMS at Mace Head. (Funny, no mention of inefficient sampling for that AMS.) Strikingly, during their "tropical marine" period OM concentrations were quite low, rather resembling our pie charts, with organics contributing 12% of the non-refractory aerosol mass for Org:SO4 = 0.16. They make no claim about what fraction of the organic material is of marine origin, but BC was ~45 ng m^{-3} and the back trajectories did pass over relatively productive waters before reaching Mace Head. It is entirely possible that none of the organics were of marine origin or that marine organics were generated up close to the sampling site. It was also noted that g(RH) tests showed particle growth rather smaller than that expected for sulfates, attributing the difference to relatively insoluble organics (presumably the 12% in the pie chart). It was further noted that other hygroscopic growth experiments in the Southern Atlantic and Indian Oceans [Maßling 2003], the Pacific and Southern Oceans [Berg et al., 1998], near Puerto Rico [Allan et al., 2008] and the Eastern Atlantic [Allan et al., 2009]. Dall'Osto et al. [2010] conclude from this that the North Atlantic is more enriched in organic aerosol than other areas.

We did not have an htDMA during VOCALS, but measured CCN concentrations agreed well with calculations from LDMA size distributions and the assumption that aerosols were ammonium sulfate [unpublished data, but we expect a paper will be forthcoming]. This adds evidence that organic material, particularly water-insoluble organic material, is not a major factor.

Zorn et al. [2008] used an AMS similar to ours in a South Atlantic and Indian Ocean cruise. While they filter their data only for ship exhaust contamination and report no soot, CO, or other pollution indicators, the OM:SO4 ratios they report are 0.07 and 0.17 for the Antarctic and South Atlantic oceans. They had much higher organics during blooms, but the associated back trajectories came over southern South America, so they were unable to exclude the possibility of continental influences.

As mentioned in our paper, there are a couple of studies that do appear to contradict our findings. Long et al., [2011] bubbled air through both productive and oligotrophic seawater and measured organic material in the resulting aerosol. Facchini et al. [2008] is similar, but used water from a phytoplankton bloom, so is less comparable with our work. They found considerable OM even when using seawater with little dissolved OC. It was present in exactly the size range we expect to see with the AMS. We don't have anything besides speculation for the difference. Maybe the North Atlantic is different. Maybe the low DOC is irrelevant if the dominant primary aerosol is water-insoluble. In any case, we look forward to seeing results from NOAA PMEL's Sea Sweep, which does a similar thing but in the open ocean rather than a lab.

The other study with results that diverge from ours is Hawkins et al. [2010], as we mention in the paper. They were on a ship in the VOCALS region at the same time as we were, but there was an unfortunate lack of instrumental overlap. They had no SP2 nor CO, which are excellent combustion indicators. They also lacked the VOC measurements present in their North Atlantic/Arctic campaign. We lacked the radon measurement they used to determine continental influence (along with particle concentration). The net effect is that it was not possible to harmonize definitions of "clean marine air". One thing to note is that the organic aerosols were split into 2 factors via positive matrix factorization (PMF). Their marine factor was dominated by carbohydrate-like material, whereas their continental, or combustion factor was dominated by alkanes. Where would biomass burning show up? With plenty of carbohydrates and a paucity of alkanes, it seems quite possible that biomass burning aerosol, would show up in the marine factor.

So to sum up, yes, there is a lot of evidence that marine organics are plentiful in productive areas, but not necessarily over the oligotrophic ocean. In fact, Mace Head appears to be a bit of an outlier. Our general results are more or less in line with other work from oligotrophic areas. What we have that's new is a rapid, accurate BC measurement that showed a much stronger relationship between BC and OM than we had expected.

The other item that's presumably contentious is our figure 10, based on the sea spray model of O'Dowd et al. [2008]. Two things struck us about that plot: it doesn't pass through 0 and since most of the ocean has very low chlorophyll-a, a small error at low Chl-a would have a very large effect on a worldwide budget. To be fair, the model was explicitly developed for the North Atlantic, not the global ocean, so that's not really a fault. However, some people have attempted to use it elsewhere, with mixed success. A simpleminded thought is that if there were no productivity, there should be no OM in the sea spray. Of course a lifespan of a few months or more would permit some to be emitted even during low productivity periods. That suggests a more complex model involving the history of the water mass might be appropriate, though beyond the scope of our paper.

We certainly admit that our discussion of that was a tacked-on afterthought. We noticed that our data were distributed along the bottom edge of the data cloud, so we added those points and hypothesized that maybe a zero intercept would work better on a global scale. We have done no modeling work and our data aren't exactly comparable with the plot (we can't separate WSOC from WIOC, which was the basis of the O'Dowd et al. [2008] model), so we can't demonstrate that our addition is actually an improvement. Such work is really beyond the scope of our paper. We have tentatively decided to eliminate that figure, since we're really not giving it a proper treatment.

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