Responses to Reviewer #2

We have restructured and added information to the Instrumentation section (Sect. 2.2). This includes a section which describes the AMS in more detail, including the exact changes made to the Squirrel fragmentation table (air fragment at m/z 29), a better description of collection efficiency and detection limits of the AMS on board aircraft, and more detailed specifics on the operation of the instrument (frequency of calibrations throughout campaigns, filter period implementation). We have added a section on the calibrations, triggering method, and detection limits of the SP2 (see specific comments below). Another section addresses the inlets, relative humidity effects and estimates of particle losses (also see below).

Answers to the reviewer's specific comments follow. The reviewer comments are in bold text, followed by our answers to their concerns. Additions/edits are reflected in a revised version of the manuscript, and are indicated by italicized font.

Reviewer wrote: "**Perhaps the most glaring issue with the paper is that the authors make extensive use of the org/so4 ratio in their analysis, but do not provide any kind of basis for this approach in the methodology. Given the mechanistic differences in the sources of both species, it must be introduced properly and the approach justified. There seems to be an implied assumption that the relative production of particulate organics and sulphate could be expected to be constant between geographical regions, but this assumption is unsafe, given the number of current unknowns associated with both production routes. There could be variations associated with all kinds of factors including biota, meteorology and atmospheric chemistry. The authors need to discuss the basis for this assumption and caveat where appropriate**."

The "implied assumption" that there is should be a constant Org:SO4 ratio isn't explicit because we assume no such thing. We emphasize Org:SO4 simply because those are the major aerosol products of the ocean, aside from seasalt itself, and often dominate the accumulation mode responsible for most CCN. Therefore we would like to know their relative importance in different areas of the ocean. In our case, we found that BC measurements forced us to find a much smaller Org:SO4 ratio than is commonly reported at high latitudes and a little smaller than in central gyre regions. Since Org and SO_4 measurements are common across all studies compared in the paper, particularly in the absence of CO and BC measurements (e.g. our TAO cruise), the ratio is a convenient and consistent way to compare and contrast fundamental aerosol properties for different geographical regions. A sentence was added to the end of the Introduction section (Sect. 1) that states: "*The use of the mass fraction of Org relative to SO_4 (Org/SO_4) will be used to illustrate that under the clean conditions established in this study, little submicron non-refractory aerosol mass can be attributed to Org. Org and SO_4 are the two largest components of submicron aerosol mass and are therefore two components that are commonly and consistently measured. Though Org and SO_4 have different production rates and chemical reactivity in the atmosphere, the ratio of Org/SO_4 is a useful and convenient tool for comparing submicron aerosol chemical composition across different geographical regions.*" The purpose of Fig. 6 and the associated analysis was intended to isolate natural aerosol (i.e., no relation to combustion indicators), and demonstrate that in those instances there was a relationship, albeit weak, between Org and SO_4, possibly indicating a common source.

Reviewer wrote: **“A related issue is the notion that organic matter and sulphate have a common source (stated at P16905, L24). The nss sulphate is known to be secondary in nature, whereas the primary vs secondary nature of the organic matter is currently a matter of debate. Even if the organics are assumed to be secondary, there is nothing to say that its precursors would originate from the same stage of a plankton bloom as DMS, be emitted in consistent relative quantities, or that SOA would be formed on the same timescale as sulphate in the atmosphere. These issues should be discussed in the context of the sulphate/organic ratios discussed above.”**

See response above.

Reviewer wrote: **“The authors should be wary of making too many implied points, especially when making comparisons with other projects. An example is on page 16906 at the end of section 4.1, where they seem to be insinuating that that a high org/so4 could be taken as indicative of the influence of biomass burning at Mace Head. In this instance, I do not consider this to be justifiable, partly for the reasons stated above but also because such a strong biomass burning influence would be very evident in the BC and CO data. In general, the authors should be more definitive about what conclusions they are making and what evidence they are basing these on.”**

The reviewer is correct that we should have been more explicit. We now argue more clearly that since BC is an aerosol product of combustion, it is an excellent tracer for other aerosol products of combustion, superior to back trajectories, overall particle concentrations, CO and other gases with different lifetimes than aerosols, and radon. (That’s not to say those other criteria are useless; BC can’t reveal continental influences other than combustion.)

BC and CO concentrations are not zero at Mace Head, and that means some of the organic aerosol found there is inevitably of combustion origin. Biomass burning influences and contamination from long range transport remain a possibility. BC concentrations are recorded as high as 70 ± 25 ng m^{-3} (Cavalli et al., 2004) during one “clean” period, a value that is over an order of magnitude higher than observed in VOCALS under clean conditions and higher than many other remote unpolluted marine values based upon similar light absorption techniques to those at Mace Head (Clarke, 1989). Low CO mixing ratios (~130 ppbv) have been observed at Mace Head that were associated with Canadian forest fire emissions (Forster et al., 2001). 130 ± 5 ppbv was the mixing ratio used to indicate background marine conditions during the Mace Head study (i.e., Cavelli et al, 2004). We do not doubt that much of the OM seen in baseline conditions at Mace Head are indeed of marine origin, as the correlation with chlorophyll-a in O’Dowd et al., (2008] and the Ceburnis et al. (2011) use of carbon isotopes demonstrate. However, one of the major conclusions of the manuscript is that given the ubiquity of long-range transport, the entire concept of background or baseline conditions is insufficient and it is necessary to either demonstrate that natural aerosols are so dominant that anthropogenic influences can be ignored or that there is a satisfactory way to isolate marine from continental aerosols. This is what this study attempts to accomplish with the use of the SP2, given its rapid response and low detection limits. The authors state as conclusion on page 161930, line 10: "*This raises questions over the appropriate choice of a clean threshold for BC used to eliminate influences of combustion aerosol when characterizing background marine aerosol*." Another sentence will be added to the manuscript to clarify this point: "*In fact, the concept of establishing background conditions is insufficient and it is necessary to either demonstrate that the natural aerosols overwhelm the anthropogenic influences such that they can be ignored, or that there is a satisfactory way to isolate marine from continental aerosols, such as the use of carbon isotopes (e.g., Ceburnis et al., 2011)*."

Ceburnis, D., Garbaras, A., Szidat, S., Rinaldi, M., Fahrni, S., Perron, N., Wacker, L., Leinert, S., Remeikis, V., Facchini, M.C., Prevot, A.S.H., Jennings, S.G., Ramonet, M., O'Dowd, C.D.: Quantificatoin of the carbonaceous matter origin in submicron marine aerosol by ^{13}C and ^{14}C isotope analysis, Atmos. Chem. Phys., 11, 8593-8606, 2011.

Clarke, A.D.: Aerosol Absorption by Soot in Remote Environments, Aerosol Sci. Tech., 10, 161-171, 1989.

Forster, C., Wandinger, U., Wotawa, G., James, P., Mattis, I., Althausen, D., Simmonds, P., O'Doherty, S., Jennings, S.G., Kleefeld, C., Schneider, J., Trickl, T., Kreipl, S., Jager, H., and Stohl, A: Transport of boreal forest fire emissions from Canada to Europe, J. Geophys. Res., 106, 22887-22906, 2001.

Reviewer wrote: **"The comparison of the organics with the SP2 black carbon under low loading conditions is very interesting, as this will not suffer from positive artefacts that beset filterbased measurements such as weakly absorbing or scattering particles. However, an unavoidable point is that the SP2 has issues of its own, particularly the detection limit with respect to BC core size (or overall scattering size, depending on the triggering method) that may decrease the overall sensitivity of the measurement, depending on the size of the particles. In this context, appropriate attention to technical differences in the instruments should be included when comparing the BC filtering methods, as this could partly explain the difference in choices of threshold values. In general, much more technical detail should be provided on the SP2 and its operation. Specifically, the detector configuration (whether PMTs or APDs were used), the calibration method used and the triggering method used (i.e. triggering on scattering or incandescence and the detection limit this entails). These details vary between units and users and could fundamentally affect the nature of the data."** and **"The authors need to cover more of the detail of the different methodologies when comparing organic data of the different studies. This isn't as much of an issue when looking at other AMS datasets, but there may be very fundamental differences when comparing with studies using offline analyses, particularly with relevance to issues such as collection efficiency and lens transmission. The same**

applies when comparing different BC methods. Ideally, this information should be included in table 1."

The detector configuration and triggering method were set up the same as DMT (found in Stephens et al., 2003 and Schwarz et al., 2006). The detector configuration was set to use PMTs for two incandescence channels (broad/narrow) and APDs employed for two scattering channels. During VOCALS, events were triggered from broad incandescence channels. The incandescence channels response was converted to refractory BC mass using a calibration curve generated with laboratory black carbon (Aquadag), sized using a long differential mobility analyzer (LDMA). The effective density of Aquadag was generated according to (Moteki and Kondo, 2010). Detection limits of 0.087-400 nm were calculated assuming a BC density of 2 g/cm^3. While it is true that significant number of BC particles may exist below the limit of SP2 detection, in the case of remote aged atmospheric soot we (and others like the NOAA and Kondo group) generally find the SP2 misses less than 10% of the ambient BC mass. If the missed mass was significant we would expect a positive intercept for regression of absorption against SP2 BC mass but this is not what we have found (e.g., McNaughton et al., 2011). It should be mentioned that very recently it has been shown by Gysel et al. (2011), that the effective density of Aquadag is lower, by about 35%, then Motecki and Kondo (2010) find. This would result in our BC measurements being too high. Even if these results are confirmed it will not change our conclusions as it does not affect the logic of our paper or the dependencies found. However, it will be mentioned in the SP2 section.

We added a paragraph in Sect. 4.4 (Sampling bias) that discusses the differences between AMS measurements and offline (filter analyses), and how the differences between these methods could affect the comparison of results.

"Not all measurements of Org compared in this study are equal. Some studies (O'Dowd et al. 2004, Cavalli et al., 2004, Quinn and Bates 2003, Phinney et al., 2006) rely on filter-based measurements of OC. Filter measurements have significant biases, including negative artifacts from volatilization of particulate-phase organics from the filter surface, and positive artifacts from adsorption of gas-phase organics onto the filter (Turpin et al., 2000). Filter measurements do not, however, suffer from the potential refractory -Org losses as the AMS does. There is also the issue of particle bounce off the collection substrate of an impactor stage during sampling, leading to inaccurate size classifications. Also, in order to convert total organic carbon (TOC) from bulk filter measurements to water soluble and insoluble organic carbon (e.g. Cavelli et al., 2004) and particulate organic matter (e.g. Quinn and Bates, 2003), TOC measurements from filters are multiplied by a conversion factor which represents a ratio between molecular mass and carbon mass. These factors, when added to the AMS biases, can contribute to possible differences when comparing studies of OM measurements. It should be noted that for the purposes of this study the term Org, which is used in the AMS community to represent the amount of POM resolved by the AMS, is used interchangeably with POM from e.g. Quinn and Bates, 2003 and WIOC+WSOC (e.g. from Cavelli et al., 2004). The same issue is true for the different methods of measuring black carbon. Filter-based techniques for measuring black carbon, such as with instruments as the aethelometer (as used in Cavelli et al., 2004), and the particle soot absorption photometer (PSAP), use a

difference in the transmission of light through a filter as it becomes loaded with aerosol. This measure of aerosol light absorption is then related to total BC through empirical formulas (Lack et al., 2008). Errors to filter-based instruments include deposit and filter matrix interactions which may change the physical and optical properties of the system, leading to inaccurate light absorption measurements, as well as the use of empirical corrections which can alter the measured change in transmission, limiting accuracies to between 20–30% (Bond et al., 1999; Virukula et al., 2005; Weingartner et al., 2003)."

Answers to reviewer specific comments:

Reviewer wrote: **"I would recommend reconsidering the title of this work as the biomass burning aspect seems to be supported by very circumstantial evidence (see below). The running title could also do with revising, given that the AMS measures organic matter (as opposed to organic carbon)."**

We disagree with the reviewer regarding the title of the manuscript. Our title does not contain biomass burning specifically. However, our Fig. 5c clearly shows for the cleaner cases there remains a strong relation between Org and BC. As the latter is only combustion derived we do not consider this circumstantial evidence. Rather, it indicates most particulate Org observed have a similar source. Hence, we see no reason to remove combustion from the title. This combustion origin is a central point of the paper and is appropriate for the title. However, "Organic carbon" in the title will be replaced with "Organic matter".

Reviewer wrote: **"The authors do not discuss what inlets are used on the different platforms. It is vital that they specify what inlet system (as distinct from the aerodynamic lens) was used for the C-130 in particular, as this can have an effect on the aerosol measured. Also, the authors must also state what humidity the particles were sampled at in each instance, as this is known to affect the collection efficiency, particularly of sea salt based particles."**

On board the C-130 a Solid Diffuser Inlet (SDI) was used for the majority of sample collection. During VOCALS the inlet was kept isokinetic during sampling by adjusting flows as flight parameters (i.e., speed, altitude, etc.) changed. Inlet losses from the SDI are most severe for large, supermicron particles (Moore et al., 2004). Moore et al. (2004) performed an inlet comparison study and found the SDI to effectively pass submicrometer (in the size range the AMS samples), as well as optically relevant coarse mode aerosol. McNaughton et al. (2007) tested University of Hawai'i's SDI against ground based measurements during the DC-8 Inlet Characterization Experiment (DICE) and found that submicrometer scattering agreed within 16%. The inlet efficiently transmits both dust and sea salt particles smaller than about 4 µm (50% cut-off) in dry diameter. Aerosol are generally dried (<60% RH) by the time they are sampled. On board the R/V *Ka'imimoana,* ¾ inch copper tubing (~30 meters) was used to bring air from the bow of the ship (forward of the stack) to the instruments housed inside the ship. The flow rate was approximately 40 liters per minute (lpm), and gravitational losses and diffusional losses for particles between 0.1 and 1 µm were estimated at < 5% (using Baron and Willeke, 2001). The sampling RH throughout the campaign was 56.15 ±

4.83 %. As mentioned earlier, this information has been added to the Instrumentation section as a new sub-section as follows:
"On board the C-130 a Solid Diffuser Inlet (SDI) was used for the majority of sample collection. During VOCALS the inlet was kept isokinetic during sampling by adjusting flows as flight parameters (i.e., speed, altitude, etc.) changed. Inlet losses from the SDI are most severe for large, supermicron particles (Moore et al., 2004). Moore et al. (2004) performed an inlet comparison study and found the SDI to effectively pass submicrometer (in the size range the AMS samples), as well as optically relevant coarse mode aerosol. McNaughton et al. (2007) tested University of Hawai'i's SDI against ground based measurements during the DC-8 Inlet Characterization Experiment (DICE) and found that submicrometer scattering agreed within 16%. The inlet efficiently transmits both dust and sea salt particles smaller than 4 µm (50% cut-off) in dry diameter. Aerosol is generally dried (<60% RH) by the time they are sampled. Five-minute filter periods were conducted a minimum of twice per 9-hour flight, usually in the beginning and at the end of flights. Filter periods during the cruise occurred once a day, and were a duration of at least 30 min.
On board the R/V Ka'imimoana, ¾ inch copper tubing (~30 meters) was used to bring air from the bow of the ship (forward of the stack) to the instruments housed inside the ship. The flow rate was approximately 40 liters per minute (lpm), and gravitational losses and diffusional losses for particles between 0.1 and 1 µm were estimated at < 5% (using Baron and Willeke, 2001). The sampling RH throughout the campaign was 56.2 ± 4.8 %."

Reviewer wrote: **"P16901, L26: The authors should account for the fact that organic matter has a lower effective density than sulphate when measured using the AMS (Cross et al., Aerosol Sci. Technol., 41, 343-359, 2007)."**

True, organic matter has a lower effective density than sulfate when measured using the AMS. However, seeing as the composition of the aerosol is mostly (>75%) comprised of sulfate, this seems a reasonable assumption for the basis of the comparison between the AMS and LDMA. The lower effective density of OM will be mentioned, and the assumption that we made when using a density of 1.7 g/cm3 will be presented. The following sentence will be added to the beginning of Sect 3: "*Although OM has been shown to have a lower effective density than that of SO_4 when sampled by the AMS (Cross et al., 2007), given the high mass fraction of SO_4 (> 75%) and the low contribution of Org (< 10 %) in the VOCALS and TAO regions, an assumption of 1.7 g cm^{-3} is justified for the purpose of this analysis.*"

Reviewer wrote: **"In the interests of fairness, care should be taken when presenting some of the other works. In some cases (albeit not that of O'Dowd and coworkers), the data presented as 'clean' is specifically within the context of the lack of local influences, with long-range transport not being discounted. In this context, presenting these works as previous characterisations of 'background marine' is slightly disingenuous."**

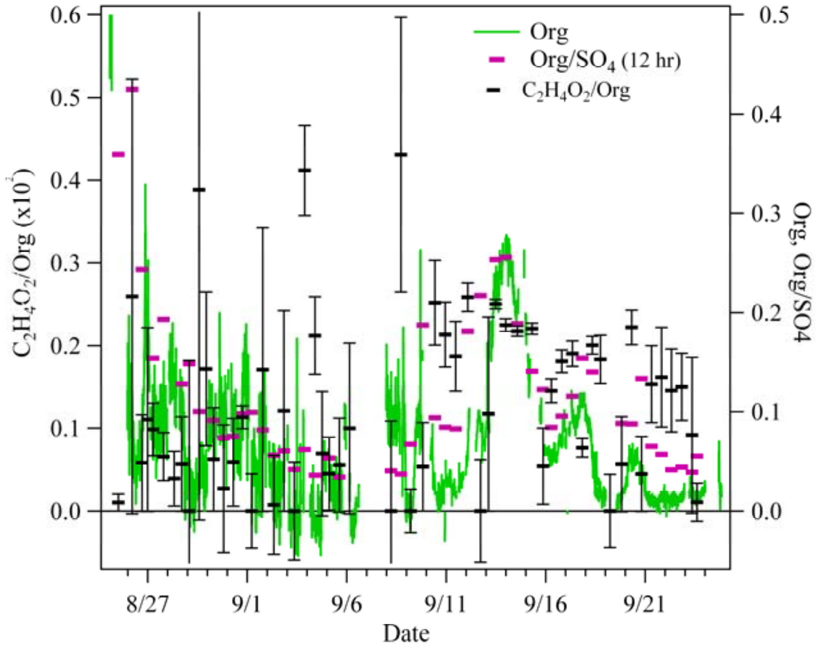
Figure 1 (along with associated text and caption) has been changed to reflect only those studies attempting to quantify "background" conditions. For example, black boxes have been removed from Clean Marine Ace-Asia, Ocean Station Papa, Trinidad Head, etc., that do not represent exclusion from long range transport. The following sentence was added to the explanation of Fig. 1 in Sect. 3: "*A distinction should be made that while some previous investigations' definition of clean aerosol focus on excluding only local sources, others (shown in bold boxes in Fig. 1) attempt to quantify a background marine aerosol for a particular region*."

Reviewer wrote: **"Given the general paucity of data in the southern hemisphere and the similarity of the techniques, Zorn et al. (Atmos. Chem. Phys., 8, 4711-4728, 2008) is notable in its omission from the comparisons. The authors should consider including this study."**

The Zorn et al., 2008 data set is now included in this study (Fig.1 and Table 1) and associated discussion.

Reviewer wrote: **"While m/z=60 can be a useful tracer, it is not unambiguous. In addition to biomass burning products such as levoglucosan, C2H4O2 can be a minor fragment of many other organic species. If the authors wish to infer the relative importance of biomass burning, they should really compile statistics on its fractional contribution to the total organics rather than the absolute signal, because I currently find figure 8 to be very unconvincing in this regard."**

Fig. 8 was reworked (see below) with the $C_2H_40_2$/Org ratio rather than the absolute concentration of the $C_2H_4O_2$ fragment.



Reviewer wrote: **"The interpretation of the Van Krevelen plot on P16908 is a little confusing. How is a high organic to sulphate ratio consistent with a non-local**

source? Also, how is a high H/C indicative of a local source? The authors should state their reasoning and assumptions more clearly."

The high O/C ratio is consistent with a non-local source (more oxygen=more aging), which happens, in this case, to be associated with high Org/SO_4 ratios. This supports the suggestion that the increase in Org/SO_4 ratio can be associated with biomass burning and/or long range transport. High H/C is indicative of a local source of carbon (less aged =less oxygenated), though high H/C ratios are primarily associated with fresh combustion. To clarify, the sentences in question were changed to: *"…i.e., the longer an aerosol is in the atmosphere, the more oxidized it will become, and the H/C ratio will decrease while the O/C ratio will increase (Heald et al., 2010). Figure 8 reveals the more aged aerosol during TAO to be generally associated with the higher Org/SO_4 ratio, indicating a non-local source for these aerosols. The higher values of Org/SO_4 are associated with higher H/C and lower C/O, suggesting a more local, perhaps oceanic, source."*

Reviewer wrote: **"The ordering of the discussion section is a little curious in that section 4.3, which deals with technical issues, occurs partway through. To me, it would make more sense to have it earlier."**

Sect. 4.3 and 4.4 deal with biases because the authors want to provide a discussion of potential issues that could affect our results (i.e., bias Org to low values). In this regard, the ordering of this section, although technical issues are addressed, seems appropriate because the majority of the section is discussion material.

Reviewer wrote: **"P16909, L14: I would contend that the 'worst case' is if the organics were externally mixed with the sulphate and had a CE of substantially less than 0.5. This could be the case if the phase of marine organics is a dry solid, in contrast to those seen in continental environments. If true, the upshot of this would be that the AMS would not measuring all of the organics present and the org/so4 ratios reported here are too low."**

While there are pathological cases like latex calibration spheres that bounce around like the rubber balls they are and are thus sampled very poorly in the AMS, real aerosols appear to be much more tractable. Laboratory (Matthew et al., 2008) and field studies (Middlebrook et al., 2011) show collection efficiencies for organic material around 45% even with solid organics on crystalline sulfates. Primary marine organics appear to be sampled well too, as the noisy but 1:1 agreement between FTIR and AMS organics during a North Atlantic cruise suggests (Russell et al., 2010). Hawkins et al. (2010), did find that small amounts of organics on dust particles were sampled poorly in an AMS, but saw no such problem with the organics they identified as marine. The following has been edited/added to this section: "*The low CE was associated with Org found on submicron dust particles originating from South America, but there appeared to be no problem detecting Org identified as marine. Besides, dust particles are of continental origin, therefore low CE values associated with Org on dust would not impact the results of this study. Furthermore, laboratory (Matthew et al., 2008) and field studies (Middlebrook et al., 2011) have shown CE for Org around 45% even with solid Org on crystalline sulfates.*

Primary marine Org appear to have been sampled well during a North Atlantic cruise (Russell et al., 2010), demonstrated by a 1:1 agreement between FTIR and AMS Org."

Reviewer wrote: **"P16909, L24: The approach of comparing the AMS with the OPC to test the presence of coarse organics is flawed. Firstly, it relies on the assumption that the organic fraction of sea salt is constant, which is not necessarily true. Secondly, if sea salt organics were present but a sufficiently large fraction was missed by the AMS (which is likely), a correlation would not necessarily be expected because it may be that a different organic particulate (with a different temporal trend) in the accumulation mode would dominate the AMS data. This would mask what little correlation was caused by the coarse-mode organics. Therefore, the lack of correlation represents nothing more than a null result in this regard."**

We were not looking for the presence of coarse organics. Instead, since both coarse seasalt particles and fine primary marine organics have the same source (bubble breaking), we sought to find a relationship between them. A relationship between sea salt Na and organic aerosol has been documented (Russell et al., 2010), but we had no direct Na measurements so looked to coarse refractory material as a proxy. We agree it is not conclusive and this is why in the previous sentence (page 16909, lines 23-24) we mention that it remains a possibility. However, we wanted to indicate that we did explore the issue. As noted above, if the OM were linked to sea-salt mass (mass mode usually 3-5 µm dry D_p) or area (area mode usually 1-2 µm D_p dry) we would expect that this would be evident as some trend with the sea-salt passing our SDI inlet and detected by our OPC (dominated by mass in 1-3 µm D_p). The absence of any relation suggests the OM seen by the AMS is not related to any OM that might vary with sea-salt mass. This is consistent with earlier observations (e.g., Fig. 2, O'Dowd, 2004) that indicated that the largest mass fraction of OM also lies in the size range well resolved by the AMS and with at most a few percent of the OM present in sizes above 1 µm. To better clarify this we have now modified the sentences in question to read: "*Hence, the potential for a significant Org fraction present on coarse sea-salt remains possible, although prior measurements of size resolved OM in marine aerosol find over 90% concentrated in sizes well resolved by our AMS (Fig. 2; O'Dowd et al., 2004). Plots and regression of Org vs. OPC coarse non-volatile mass (a sea-salt surrogate – as used in the discussion of Fig. 2) also revealed no evidence of a relation between the OM concentrations measured by our AMS and sea-salt concentrations.*"

Responses to technical corrections:

Reviewer wrote: **"Throughout the manuscript, the authors refer to organic matter as 'Org'. It would be much better to use 'OM', as this is a more universally used term."**

We argue that while OM is more universally used, the AMS community uses the abbreviation "Org" to refer to the OM resolved by the AMS. In this paper, OM vs Org forms a useful distinction, as there is possibly OM too refractory to be detected as Org.

P16898, L1: Technically, the role of natural aerosols in the earth's climate should not be described as a 'forcing' if the IPCC definition is used. Suggest changing to 'processes'.

The term 'forcing' was replaced with 'processes'. The sentence now states: *"The role of sea-salt aerosol and non-sea-salt sulfates in climate processes…"*

P16898, L27 (and elsewhere): 'Scavenging' is often used to describe specifically the process by which interstitial black carbon is incorporated into cloud droplets. Wet deposition as a whole involves other mechanisms in addition to scavenging (activation and washout), so I would suggest using a more general term to describe the loss mechanism.

Changed instances of 'scavenging' to 'removed'. i.e., p16898, L27 now states:

"However, unless aerosol from sources upwind have been effectively removed by boundary layer precipitation, these sites remain subject to potential influences from local and/or long range transport."

P16900, L12: Drewnick et al. (2005) is an inappropriate reference for the HR-TOFAMS, as this described the earlier C-TOF-AMS. Suggest DeCarlo et al. (2006) or Canagaratna et al. (Mass Spectrom. Rev., 26, 185-222, 2007) instead.

The following references were added both in text, and in the references section:

Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch, T. B., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L. R., Trimborn, A. M., Northway, M. J., DeCarlo, P. F., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Chemical and microphysical characterization of ambient aerosols with the Aerodyne aerosol mass spectrometer, Mass Spectrom. Rev*.,* 26, 185-222, doi:10.1002/mas.20115, 2007.

DeCarlo, P.F., Kimmel, J.R., Trimborn, A., Northway, M.J., Jayne, J.T., Aiken, A.C., Gonin, M., Fuhrer, K., Horvath, T., Docherty, K.S., Worsnop, D.R., and Jimenez, J.L.: Field-deployable, High-Resolution, Time-of-flight Aerosol Mass Spectrometer, Anal. Chem., 78, 8281-8289, 2006.

P16900, L25: The specific changes made to the fragmentation table should be stated.

The following paragraph was added to the AMS description (now Sect. 2.2.2):
"The fragmentation table in SQUIRREL was adjusted to give zero Org mass concentrations during filter periods. This was achieved by altering the air mass fragment coefficient at m/z 29 (frag_air[29]). The default coefficient is used to represent an isotopic factor and describes the relative amount of $N^{15}N$ *at m/z 29, which is related to the signal of* N_2 *at m/z 28. However, depending on the threshold setting and possible saturation effects, this isotopic factor may be slightly different than predicted, and may*

be multiplied by a factor, close to 1. In our case, the isotopic factor was adjusted up to give zero Org signal at m/z 29 during a filter period.”

P16908, L9: Do the authors mean ‘lower’?

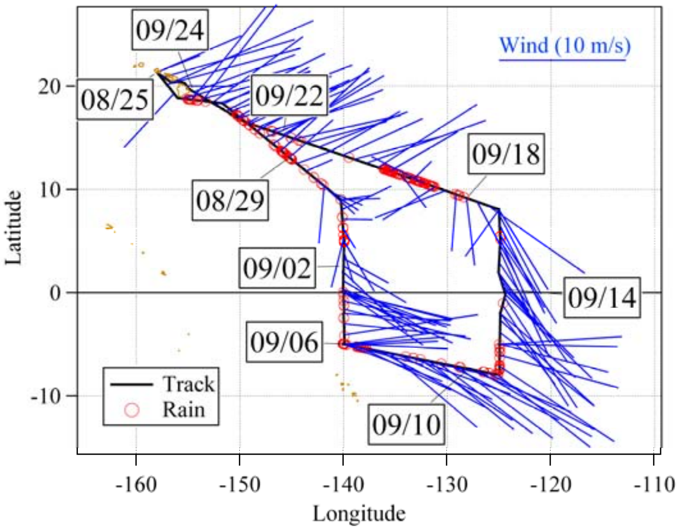
The authors meant lower. The sentence has been changed to: *“Because the signal to noise level is lower for the* $C_2H_4O_2$ *peak, error bars (1σ) are shown as well.”*

P16922: Allan et al. (2004) used a VOC tracer (MTBE) to screen for local influences, not back trajectories.

Table 1 has been changed to reflect Allan et al., 2004 used a VOC tracer to screen for local influences.

P16926: I feel that having the dates at angles makes the figure difficult to read.

An updated version of Fig. 4 is below.



Additional references were added to the manuscript:

Baron, P.A. and Willeke, K. (editors) Aerosol Measurement: Principles, Techniques and Applications (second edition), 37 chapters, 1144 pages, John Wiley & Sons, 2001.

Bond, T. C., Anderson, T. L. and Campbell, D.: Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols, Aerosol Sci. Technol., 30, 582, 1999.

Ceburnis, D., Garbaras, A., Szidat, S., Rinaldi, M., Fahrni, S., Perron, N., Wacker, L., Leinert, S., Remeikis, V., Facchini, M.C., Prevot, A.S.H., Jennings, S.G., Ramonet, M., O’Dowd, C.D.: Quantificatoin of the carbonaceous matter origin in submicron marine aerosol by ^{13}C and ^{14}C isotope analysis, Atmos. Chem. Phys., 11, 8593-8606, 2011.

Clarke, A.D.: Aerosol Absorption by Soot in Remote Environments, Aerosol Sci. Tech., 10, 161-171, 1989.

Cross, E. S., Slowik, J. G., Davidovits, P., Allan, J. D., Worsnop, D. R., Jayne, J. T., Lewis, D. K., Canagaratna, M., and Onasch, T. B.: Laboratory and ambient particle density determinations using light scattering in conjunction with aerosol mass spectrometry, Aerosol Sci. Technol., 41, 343-359, 2007.

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