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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol composition and sources in the Central Arctic Ocean during ASCOS" *by* R. Y.-W. Chang et al.

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The paper is a valuable and interesting contribution to aerosol composition over remote oceanic regions and especially over the unique Arctic Ocean. Authors have used modern tools to make source apportionment of organic matter and used a number of elemental tracers to facilitate principal source apportionment. The manuscript is suitable for publication in ACP subject to addressing many of the important issues when interpreting the results.

Authors have missed a recent but very important paper detailing biogenic marine organic matter signatures using high resolution AMS by Ovadnevaite et al. (2011). This paper is the most detailed describing marine organic matter spectroscopic features and interpreting AMS mass spectra. Even though Chang et al. used C-ToF AMS in their study it is even more important to take into account considerations from a high resolution instrument. In any event authors should make a better effort at interpreting PMF fragmentation pattern by comparing with available references when attempting to conclude source origin. Oxidation factor is just one part of a story even more so that the parameter is not measured having C-ToF. Referring to the highly oxidised nature of primary biogenic OM over the Arctic Ocean it could be its typical feature as claimed by L. Russell group (with references included and briefly discussed in the paper). Also study by Zorn et al. (2008) should be considered in the paper as being the first detailed paper on MSA detection by AMS and presenting extensive measurements of oceanic sulphate/MSA.

We thank the reviewer for thoroughly reading our paper and for their insightful comments. We were unaware of the paper by Ovadnevaite et al. (2011) and we thank the reviewer for bringing it to our attention. References to this paper as well as the work by Zorn et al. (2008) are now included. While we agree that comparing factor mass spectra with reference mass spectra is useful in understanding their sources, the source of a factor should only be confirmed with a correlating time series (Ulbrich et al., 2009). As such, it is difficult to attribute a single source to the Organic factor. This will be further discussed below.

Specific comments as follow:
Abstract I believe that the last sentence of the abstract may change when the authors reconsider their results and interpretations. In my opinion the Organic-rich Factor has little to do with continental aerosol (including biomass burning), but rather is a signature of processed primary or oxidised marine biogenic aerosol, not surprisingly non-correlating with any other factor or inorganic species. Consequently, Organic-rich Factor most likely is Marine Biogenic

Factor II. Considering that the ice breaker spent significant amount of time over pack ice (far from open ocean), contribution of truly primary biogenic marine factor could have been limited while at the same time there was ample time for oxidation processes to take place or OM was produced in highly oxidised form in the first place according to Russell et al. (2010). It is simply inconceivable that Organic-rich Factor contained continental OM but none of the other ubiquitous species like nssSO4, NO3, BC or elemental tracers like 222Rn and 210Pb or selected VOCs. These considerations will be repeated below as appropriate.

The Organic Factor does correlate to some degree with radon, suggesting a possible continental contribution (see newly added Figure 11), although the BC data are unavailable at this time and we cannot make any comparisons. In addition, pure organic aerosol have been observed at continental sites that are in forested regions. As such, it is possible that there are continental contributions to this factor. On the other hand, the mass spectrum of this factor resembles that of Frossard et al. (2011); Ovadnevaite et al. (2011) and sugar standards from Russell et al. (2010), pointing to possible marine sources. As discussed in more detail below, although it would be nice to attribute this factor to only one source, we feel that the source of the Organic Factor cannot be identified with 100% confidence at this time and we have instead expanded the discussion to include various possible sources such as both primary and secondary marine sources, continental sources as well as biomass burning sources. In response to the reviewer's comments, the possibility of a primary marine source has been emphasised in the abstract and conclusions.

P14840. Line 13. Suggest using "aerosol number concentration" or "mass loadings".

P14843. The last paragraph should be moved above the previous paragraph.

These have been changed in the revised manuscript.

P14844. Inlet issues. Please calculate and state flow conditions in the inlet with Reynolds numbers. Tube and particle Reynolds numbers should be presented for the main (∼17000Re) and AMS inlet (seems to be laminar at ∼1100Re). Therefore, the main inlet was not laminar, thus facilitating loses, even of particles as small as 1umn diameter and of all larger ones. Even though AMS aerodynamic lens do not transmit particles above 1um, some of the larger submicron particles could have been lost as well. Considering internally mixed Org and sea salt (SS) in sea spray significant amount of organic matter could have went undetected. It is also important to mention whether AMS was isokinetically sub-sampling from secondary inlet or sampled at 100cc/min directly. Assumption of <30%RH is very arbitrary as aerosol particles often require long equilibrium time (much in excess of 5s as was the case in this set-up). Please refer to (Chan and Chan, 2005) as a reference of ongoing debate in HTDMA community. Therefore, it could well be possible that RH did not drop below 40% which is approximately an efflorescence point of sea salt leaving particles wet (which is nothing wrong by itself but has implications). If particles were dry as authors claim then internally mixed sea salt and primary organics could have been possibly lost due to sea salt bounce off the vaporiser.

Based on the reviewer's arguments, the chances of detecting sea salt particles in the AMS are very slim. If they were wet, then they would likely be too large to be transmitted through the instrument. However, if they are dry, then they might be transmitted into the AMS, but would bounce in the oven and go undetected. The flow in the main sampling inlet was kept turbulent such that the air sampled by the numerous isokinetic secondary lines would be well-mixed. In response to the reviewer's comments, the

Reynolds numbers are now included in the manuscript and the sentence referring to the RH has been revised to:

"The room temperature was at least 20 K warmer than ambient, resulting in an RH of $<$30% in the lines. It is possible that the aerosol did not have sufficient time to effloresce, shifting the range of dry particle diameters sampled by the AMS to smaller sizes."

P14845. Lines 19-21. Flow rate of AMS is 100cc/min, not s-1. Authors state that filter measurements were taken twice a day, but were they actually used in data processing? Possibly not, as Figure 2 shows negative concentrations. Otherwise explain negative concentrations.

We thank the reviewer for pointing out the mistake in the unit for the inlet flow. This has been corrected in the revised manuscript. In regards to the filter measurements, if they had not been used to correct the air peaks of the mass spectrum, the data would have been much more negative than the presented time series. Given the low mass concentrations measured during this study, we feel that $\sim$ 90 minutes of organic mass concentrations $<$ -0.03 μg m^{-3} was an acceptable fraction over the 6 week period. The negative organic mass concentrations were mostly due to the subtraction of a very noisy ammonium signal, especially at *m/z* 15.

Lines 15-20. How MSA was actually quantified to derive SO4/MSA ratio? If a factor was used to multiply m/z79 or 96 then authors should know that MSA fragmentation pattern is temperature and, therefore, instrument dependent according to Zorn et al. (2008). How is then the factor applicable to the C-ToF which authors used when vaporiser temperature calibration is a significant

problem in AMS.

The fragmentation pattern of Langley et al. (2010) depend on both *m/z* 79 and 96. The reviewer makes a valid point that these peaks are temperature dependent. However, the instrument used in this study was the same as that used by Langley and co-workers and the nominal temperature was the same. As such, the fragmentation pattern used should be valid for our study. This discussion is now included on Page 8 line 9.

P14846. Line 2. Please state cut-off range of the impactor. In fairness, cascade impactors are unreliable for this purpose due to independent inlet, sampling regime, etc. Collection efficiency should normally be used as 0.5 unless calculated composition dependant CE according to Middlebrook et al. 2011 (http://cires.colorado.edu/jimenez/group_pubs.html)

The second lowest stage of the impactor had a cut-off of 665 nm while the third lowest stage had a cut-off of 2.1 μm. Collection efficiencies calculated when comparing MSA and SO_4^{2-} measured by the AMS to either of these cut-offs ranged from 0.5 to 1.0 depending on the statistic used (median, geometric mean, arithmetic mean). While we believe that in-situ measurements should over-ride conventional approaches, we agree that using a collection efficiency of 0.5 will give our results the same uncertainties as other reported AMS measurements and the numbers have been changed to reflect this. In addition, the following sentence has been added to Page 8 line 26:

"However, since the uncertainties were large, a standard collection efficiency of 0.5 was used such that our results have the same assumption as other AMS studies."

P14851. Lines 1-5. Authors claim about absence of ssSO4 is true if particles were completely dry (which authors assumed but not measured). Assumption about refractory nature of sea salt is overstated in AMS community. If sea salt particles were partly wet, sea salt could have been partly vaporised as NaCl molecules (in the end m/z58 and 60 were present in spectra). Authors could check the ratio of m/z58:60 and if it was close to 3:1 that is a signature of SS (m/z 60 is Na37Cl isotope molecule). If authors had high resolution instrument this issue would have been easy to check. I was wondering if C-ToF signal can resolve m/z58 fine structure. Was the signal clean Gaussian or slightly distorted as over the oceans with polluted background (including ship emissions) m/z 58 should split between NaCl signal (m/z 57.95 and several organic fragments slightly above 58). In any event ssSO4 would have been a minor component.

It is true that a high resolution instrument could have easily resolved this issue, however we did not have one on board. Figure 1 of this response shows the total sulphate and non-sea salt sulphate (as determined from chloride measurements) from the lowest two and three stages of the cascade impactors (25-665 nm and 25-2120 nm vacuum aerodynamic diameter, respectively, at 50% RH). Since the particles collected by the cascade impactors had an RH of 40–50% (monitored throughout the cruise), submicron particle loss from bounce would be expected to be less than in the AMS and most representative of the ambient submicron aerosol. All of this would suggest that the contribution of sea salt sulphate was negligible for submicron aerosol.

In addition, the *m/z* 58 to *m/z* 60 ratio for the Marine Biogenic factor was 2.4 while that for the Continental and Organic factors were 2.0, suggesting that this signal was not due to NaCl and that sea salt was not sampled by the AMS. For comparison, the ratio when bubbled sea water was sampled by the AMS during the cruise was 3.0 - 3.1, as would be expected from the isotopic ratio of chlorine. In response to the comments from both reviewers, this sentence now reads: "...based on cascade

impactor measurements, submicron sea salt sulphate concentrations were negligible".

P14851. Lines 5-18. It is enough stating that values were in agreement with other studies and refer to the Table which is way much clearer.

This paragraph has been shortened and the reference to Table 2 emphasised.

P 14852. PMF results. In general PMF results are trustworthy with the presence of J. Paatero in the author list. However, authors should do a better job of describing and discussing spectrometric features of the factors obtained in their analysis with those of other appropriate studies, especially of Ovadnevaite et al. (2011) in case of Marine Biogenic and Organic rich Factors.

Marine Biogenic Factor must be compared in detail with aforementioned study.

This is now included in the newly added Table 3 and discussion included in Sect. 3.2.1.

P 14855. Lines 14-17. Just the presence of high pressure is not an evidence of mixing from aloft (does it mean free troposphere?). I think the entrainment has been too often a waste bag of explaining anything unusual.

In response to the concerns of both reviewers, this description has been expanded to the following:

"This was a period of recoupling (27 and 29 August) and decoupling (28 August) of a shallow (~ 100 m deep) surface-based mixed layer with the upper part of the boundary layer - the upper part of which contained stratocumulus clouds. This recoupling can be clearly identified in radiosonde profiles, and turbulence profiles from a tethered balloon. We speculate that this surface air mixed with the upper part of the boundary layer was influenced by continental combustion (average acetonitrile mixing ratio of 0.080 pptv compared to campaign-long average of 0.048 pptv) and high particle masses."

The intent was to describe the meteorological conditions since direct proof of transport was unavailable.

Line 19. Please state 222Rn concentrations. I am confused how 210Pb is a better tracer of continental source having half-life of 22.3y when 222Rn has only 3.8d. Clearly, Rn should be more sensitive of contact with land. Even if there was no contact with land for 4days, only half of Rn would have decayed, excluding wet and dry deposition, but this would have equally affected 210Pb as well. Authors should provide with numbers of the elemental tracers, not just correlation coefficients. Please refer to the Biraud et al. (2000) when considering Rn concentrations.

A variation of Figure 2 of this response, which shows the ^{222}Rn for the entire study and the Organic Factor averaged to a similar time scale, has now been added to the manuscript. It is because of the lower correlation of the Continental Factor with ^{222}Rn compared to ^{210}Pb that we conclude that the air must have last been in contact with land for more than a week, since a mere 4 days would have still produced a moderate correlation. In fact, ^{222}Rn appears to be anti-correlated to the Continental Factor from 36-30 August, when the Continental Factor is the highest. Spurred by the reviewer's thorough reading, we realised that the correlation coefficient for radon presented in

the original manuscript was incorrect and the correct value (0.15) is now included.

Organic Factor spectrometric features should be more carefully examined as it could also be Marine Biogenic Factor II only of processed or oxidised primary marine organic matter.

P 14859. Last paragraph of the conclusions should be reconsidered after re-examining the data.

Based on the correlation at the end of the study of this factor with radon and the lack of a strong sea salt signal in the mass spectrum, it is difficult to attribute this factor completely to marine sources. While the mass spectrum of a factor can resemble a specific source, a correlating time series is needed to identify a source. In light of this, we have expanded the discussion in Sect. 3.2.3 to include various possible sources of the Organic factor and the evidence for and against each. In addition, in response to the reviewer's comments, we have emphasised the possibility of a primary marine organic source in the abstract, Sect. 3.2.3 and the conclusions.

Table 2. It would be more appropriate to present geometric mean (median) and the range of sulphate and MSA concentrations from this study and not "below value" which implies that measurements were very uncertain or below detection limit.

The median is included in Table 1. The ranges for this study have been changed to $\leq$ the detection limit to the maximum.

Figure 2. I am curious what happened on August 19th to 20th when some peculiar OM plume has been missed as can be inferred from MSA and Org mass concentrations right after the measurements have resumed. It is interesting why Figure 7 does not have that gap with Continental Factor time series uninterrupted?

Unfortunately, the detector of the AMS was replaced during that time and the instrument did not measure the interesting event. Because the resolution of the ^{210}Pb measurements was 24 hours, the AMS measurements were averaged over this time. The average preceding this interruption had 15 hours of measurement while the one proceeding the interruption was averaged over 6 hours.

Figure 3. Caption should clearly state that factors consider all species fragments, not only those of organic matter.

This has been changed in both Figures 3 and 4.

Figure 4 and 8.
This comment has been broken up so that our responses can be clearer.

This is really intriguing and confusing. Marine Biogenic Factor suggests secondary features of chemical species with prominent sulphate and MSA fragments (could it be marine biogenic secondary factor?).

Since both MSA and sulphate are oxidation products of SO_2, we are implying that at least the inorganic part of this aerosol is secondary in nature. The word "secondary"

has now been included in the text on line 4 of Sect. 3.2.1.

However, fragmentation pattern of the same Marine Biogenic Factor only performed on OM mass, demonstrates features of primary marine organic matter presented by Ovadnevaite et al. (2011): m/z 41, 43, 55 and also 58 and 60, most likely sea salt (if the ratio 3:1 holds).

As discussed above, the ratio of textitm/z 58 to 60 is 2.4 and is therefore unlikely to be sea salt. Nevertheless, the organic component of the Marine Biogenic Factor does correlate best with the primary organics measured at Mace Head, although not so well with sugar standards such as glucose and manitol measured by Russell et al. (2010) (see the Table 5 newly added to the manuscript). The last paragraph of Sect. 3.2.1 has been expanded to discuss this further.

At the same time Organic-rich Factor in Figure 4 shows features of primary biogenic organics, but same factor in Figure 8, exhibits largely oxidised character of OM (note dominance of m/z 28 (CO) and 44 (CO2) as in Ovadnevaite et al. (2011) and consistent with L. Russell papers noting polysaccharides and carbohydrates) which prompts to the author's conclusion about aged continental OM (including biomass burning).

Thanks to the reviewer's careful observation, we realised that the mass spectra for the Continental Factor and the Organic Factor were mis-labelled and have corrected this in the new version of the manuscript. Both the Continental Factor and the Organic-rich Factors have prominent *m/z* 28 and 44 peaks. However, it should be noted that in the fragmentation table for a unit-mass resolution AMS, the signal at *m/z* 28 is assumed to be equal to *m/z* 44 (see Aiken et al. (2008)). As such, the high signal at *m/z* 28 is only inferred as opposed to the study by Ovadnevaite et al. (2011) where it was measured directly with a high-resolution mass spectrometer. Nevertheless, the high signal at *m/z* 44 shows the oxidised nature of the organic regardless of its origin.

However, that same Organicrich Factor does not contain characteristic anthropogenic fragments 57 and 71 (only 43, which is difficult to apportion without high resolution AMS, i.e. oxidised or hydrocarbon).

The mass spectrum referred to by the reviewer is in fact the mis-labelled Continental Factor. Low signals at textitm/z 57 and 71 are not that unusual for LV-OOA which have been formed over continental sources (e.g. Zhang et al., 2005).

If Organic-rich Factor has anything to do with continental aerosol it must correlate with elemental tracers like BC, 222Rn or 210Pb. But it does not seem so as only Continental Factor correlates with 210Pb, and rightly so (however, why not with Rn as mentioned above). I would suggest to plot Marine Biogenic and Organic-rich Factors together (and along with 210Pb/222Rn) to see when they coincided and when diverged.

In response to the reviewer's comment above, the newly added figure shows that radon does correlate to some degree with the Organic-rich Factor, especially at the end of the study. However, the return of the *Oden* to open waters could also explain increased organic aerosol from marine sources. At this point, we feel that it is difficult to attribute the Organic Factor to a single source and we have instead expanded the description for this factor in the revised manuscript to discuss various possible sources (marine sources, secondary continental and biomass burning) and the evidence for and against them.

In addition, as requested by Reviewer 1 and to address the concerns of Reviewer 2, the correlation coefficient of all the factor time series and mass spectra have now been summarised in a new table. This includes a comparison with the mass spectrum of Ovadnevaite et al. (2011) and Frossard et al. (2011), which shows that there is good correlation between the organic components of the Continental and Organic Factors, and slightly lower for the Marine Biogenic Factor and is now discussed in Sections 3.2.1-3.2.3.

Figure 5. Marine Biogenic Factor and DMS relationship is interesting, but can be further explored considering DMS and solar radiation relationship (Vallina and Simo, 2007). It may reveal secondary as well primary character of the Factor if possible to split it based on DMS measurements.

A more thorough study of the sulphur budget during this cruise is warranted (e.g. Nilsson and Leck, 2002), but would be beyond the scope of this paper. Since both sulphate and MSA, which together make up 80% of the Marine Biogenic Factor, are secondary products of DMS oxidation, this aerosol must have secondary sources. It is only the nature of the organic component in this factor that is in question. Relating DMS to shortwave radiation as in the study by Vallina and Simó (2007) can help explain the secondary nature of the aerosol, but would not identify the nature of the organic component.

Figure 9. To claim any influence from biomass burning events, AMS spectrum should contain significant fragment at m/z60 (levoglucosan) or other typical biomass burning fragments (refer to Jimenez group papers at the link above), but it does not seem to be the case. In any case BB events happened to be too far away to have conceivable influence on aerosol chemical composition at the measurement point of thousands of kilometres away. Again elemental

tracers like CO, BC, 222Rn, 210Pb or selected VOCs should give an indication. For instance, Canadian forest fire signature via CO was discerned in Ireland 5000-7000km away (Forster et al., 2001) while aerosol was largely removed by wet and dry deposition during transport. Quite contrary, 210Pb concentration was among the lowest on September 4-5th (what was 222Rn?), also August 20-21th when Organic-rich factor was high in Figure 7. Unless authors have a better direct evidence of BB plume or its residue detection it is suggested to stay away from such a general and speculative claim and interpret what is at hand: AMS fragmentation pattern and PMF. Authors obtained a very interesting dataset which is suggesting intriguing conclusions; therefore, they do not need to resort to generalised claims to comply with every possible aerosol source.

Laboratory studies (Grieshop et al., 2009) and field measurements (Capes et al., 2008) have shown that the signal at *m/z* 60 can decrease as biomass burning aerosol are aged (see panel (d) in Figure 3 of this response taken from Capes et al. (2008)). Both of these studies suggest that as biomass burning aerosol are aged, the mass spectra approach that of OOA, where the signal at *m/z* 44 dominates.

Unfortunately, CO was not measured and the BC data are not yet available so comparisons with these data cannot be made. It is true that acetonitrile levels are not high during the biomass burning event identified by FLEXPART, however, radon concentrations were elevated. As discussed in the previous point, we feel that at the current time, the origin of the Organic-rich Factor cannot be positively identified and as such, we prefer to discuss various possible sources and the evidence for and against each of them.

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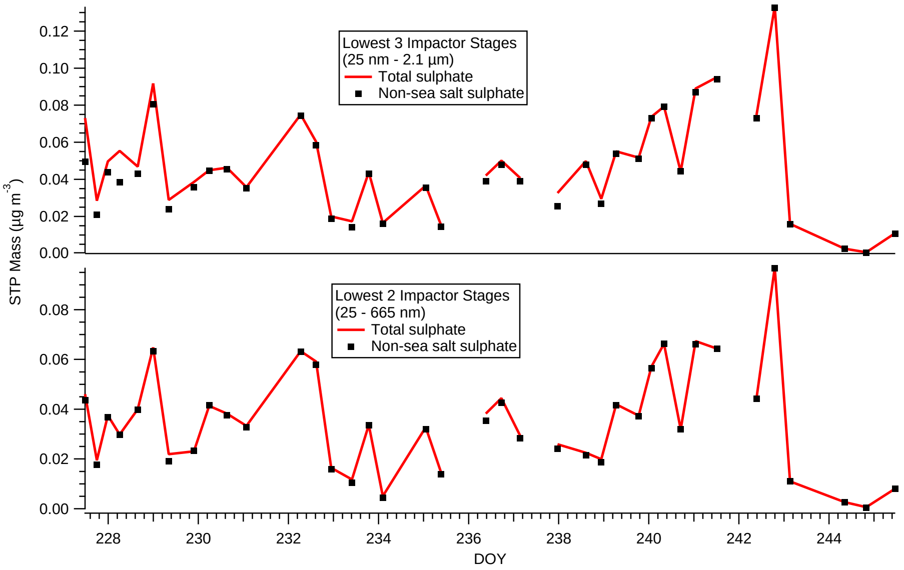

Fig. 1. Sulphate from the lowest two and three stages of the berner cascade impactors.

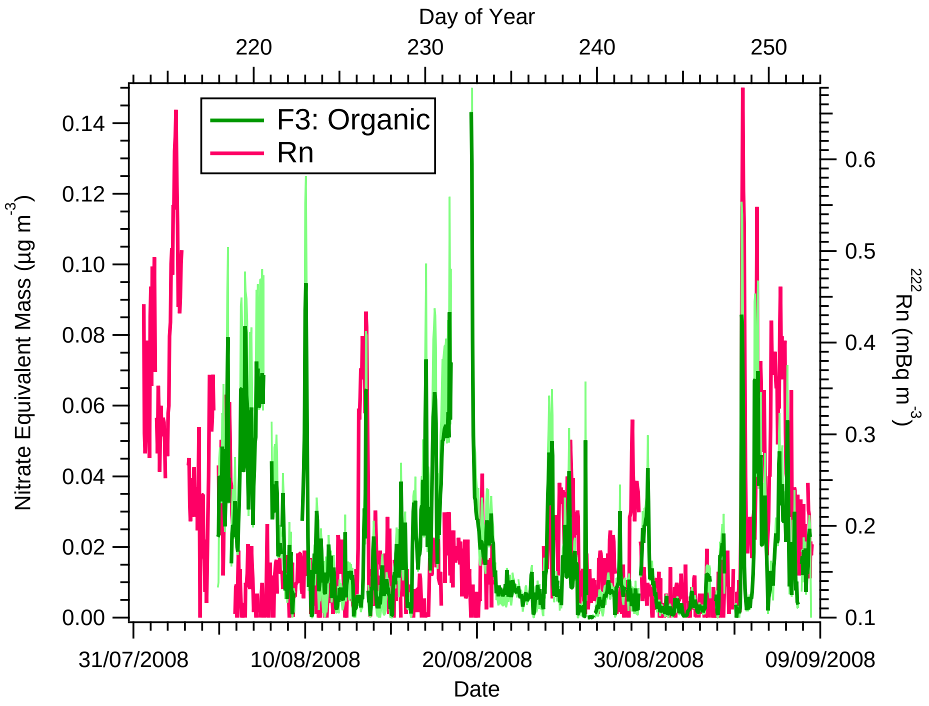

Fig. 2. Time series of Organic factor and radon.

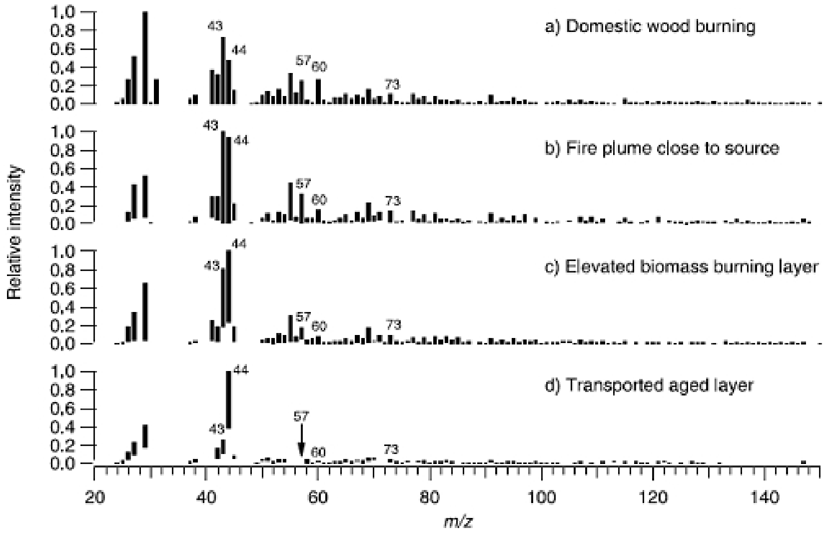

Fig. 3. Fresh and aged biomass burning aerosol over West Africa from Capes et al. (2008).