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Title: Measurement report: Aerosol vertical profiles over the Western North Atlantic Ocean during the North Atlantic Aerosols and Marine Ecosystems Study (NAAMES)

Responses to Anonymous Referee #1

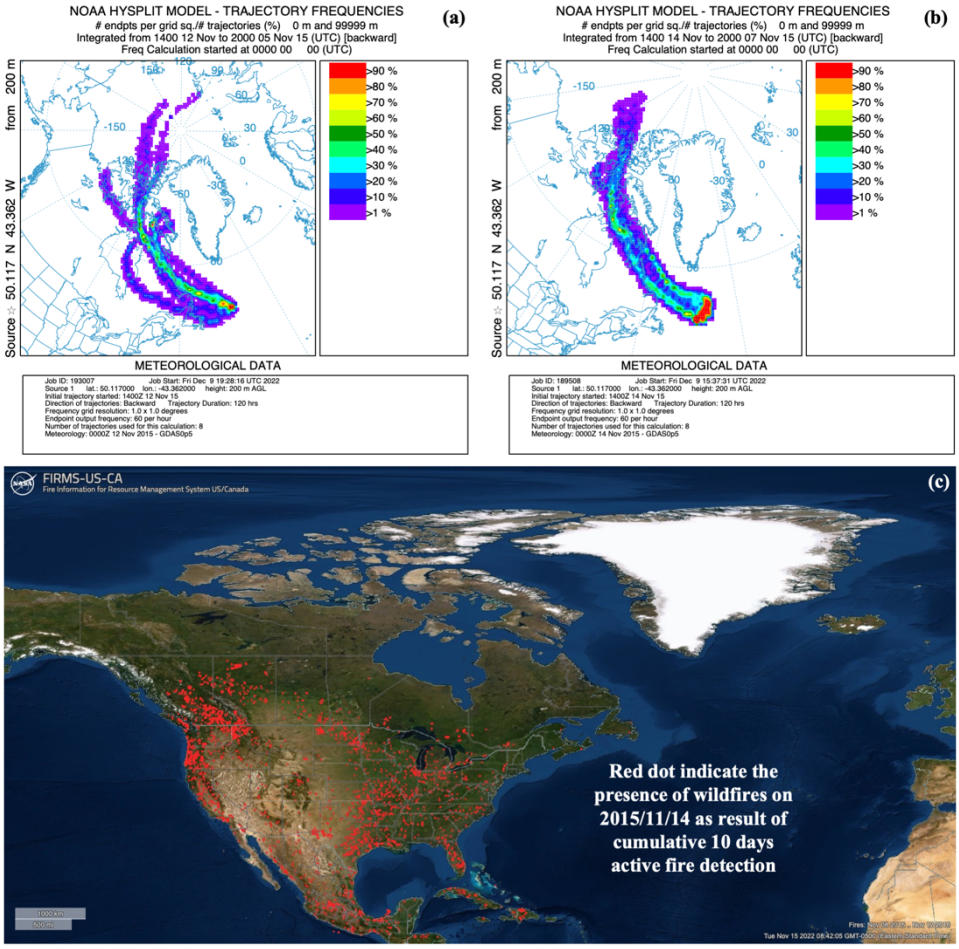
Comment: *The measurement report by Gallo et al. analyzed in-situ measurements during three NAAMES field campaigns in winter, spring, and summer, respectively. The results are interesting and well discussed, and I only have a few concerns.*

Response: We thank Anonymous Referee #1 for his/her support of our work. We have revised the manuscript according to your suggestions. All the alterations to the manuscript are shown in the track changes revised version of the manuscript included within this document. Please find our point-by-point responses below - the original Referee #1 comments are highlighted in italic black and our responses follow in blue. (blue).

Detailed Comments:

1. Sect 3.2*: During NAAMES 1, why only BC show the MBL peaks on 12th, 2015 and Nov. 14th, 2015? If it was the continental origins, the CO should also be higher. Has the authors considered the possibility of ship emissions?*

Response: We thank Anonymous Referee #1 for this suggestion. We have conducted further analysis to constrain the possible origin of enhanced BC levels in the MBL on November 12th and 14th, 2015. First, we have assessed 5-days Hysplit frequency backtrajectory (Fig. a, b - below) and we evaluated the potential presence of wildfires along the air masses transport path using NASA Worlview FIRMS VIIRS Fire and Thermal Anomalies product (Fig. c below). The images below show that air masses originated over the Artic and did not intercept wildfires prior the arrival to the NAAMES domain. The influence of high levels of pollution along the air mass trajectories from the Artic to the Western North Atlantic is also unlikely.



We compare our results to the BC measurements simultaneously collected on the research vessel *R/V Atlantis* through a Single Particle Soot Photometer, and we find elevated concentrations of BC (> 500 ng m^{-3}) at the surface ocean on November 12th and 14th, 2015. These analysis and the low level of CO previously observed, confirm Referee#1's hypothesis that possibility attribute the BC peaks in the MBL to ship emissions. Therefore, we have revised the manuscript (**page 6, line 38** of the track changes revised version of the manuscript) and added the figure in the supplemental materials (**Fig. SI.2** of the track changes revised version of the manuscript):

> During NAAMES-1, we observe a mean CO concentration = 87.2 ± 6.6 ppbv and no significant variations within days. CO vertical profiles show similar concentrations at all the altitudes sampled, being only slightly higher in the MBL (mean = 90.1 ± 3.3 ppbv) than in the FT (mean = 86.1 ± 7.2 ppbv) ~~(Fig. 3a, Fig. SI.1.a, and Fig. SI.2.a)~~ (Table 3, Fig. 3a, Fig. 4a, and Fig. SI.1.a). BC concentrations are also generally low ~~(mean of 6.2 ± 11.2 ng m^{-3})~~ ~~(mean of 8.3 ± 21 ng m^{-3} in the MBL, and 5.1 ± 4.8 ng m^{-3} in the FT)~~, characteristic of unpolluted conditions observed during the winter (Table 3, Fig. 3b, Fig. 4b, and Fig. 6a). The only exceptions are Nov. 12th, 2015 and Nov. 14th, 2015 when BC exhibits peaks in the MBL up to 101.4 ng m^{-3} and 106.2 ng m^{-3} respectively, at altitudes < 1500 m ~~(Fig. 3b, and Fig. SI.2.a.1-3)~~ (Fig. 4b, and Fig. SI.2.a.1-3). The enhanced BC levels might be the result of ship-traffic emissions. Analysis of 5-days Hysplit frequency backtrajectory indicate the arrival of air masses from the Artic suggesting clean maritime influence (Fig. SI.2.a-b). Furthermore, Worlview FIRMS VIIRS Fire and Thermal Anomalies products show absence of wildfires on the air mass transport path prior the arrival to the NAAMES-1 domain, therefore excluding the contributions from biomass burning to the BC level observed (Fig. SI.2.c). After removing the BC measurements affected by ship-traffic emissions (data discarded altitude < 1500 m on Nov. 12th, 2015, and altitude < 1050 m on Nov. 14th, 2015) we found mean BC concentrations in the MBL of 1.8 ± 0.9 ng m^{-3} and lower than in the FT. ~~the intrusion of polluted air masses from continental origins due to wintertime low-altitude long-range transport.~~

~~Field studies conducted in several locations over the North Atlantic ocean region have shown background concentrations of BC ranging between 10 and 40 ng m^{-3} under unperturbed marine conditions (O'Dowd et al., 2004; Shank et al., 2012; Pohl et al., 2014; Cavalli et al., 2016), while BC concentration up to 600 ng m^{-3} have been reported during periods of time affected by intense continental emission plumes (Corrigan et al., 2008).~~ Non-refractory aerosol chemical composition analysis also reveals low concentrations of sulfate (0.05 ± 0.03, 0.03 ± 0.03, and 0.07 ± 0.03 µg std m^{-3} respectively at the surface ocean, in the MBL, and in the FT) ~~(Fig. 4a, Fig. 5b, and Fig. SI.2.a)~~ (Fig. 3c, Fig. 5a, and Fig. 6b), and organic (0.1 ± 0.07, 0.05 ± 0.08, and 0.08 ± 0.06 at the surface ocean, in the MBL, and in the FT, respectively) aerosol mass ~~(Fig. 4b, and Fig. SI.1.b, and Fig. SI.2.a)~~.

2. Fig. 6a-c: *The correlations between CO, Sulfate, Organics and BC during NAAMES 1 are strongly biased by the BC outliers. How would the correlations be like if any BC > ~20 ng/m3 are excluded, and what's the explanations? That would be more representative of the pristine marine environment in winter. ?*

Response: We evaluate the concentrations of BC on November 12th and 14th, 2015 during the periods in which measurements were likely affected by ship emissions and we found BC being > 18 ng m^{-3}. We agree with Referee #1 that the BC measurements affected are not representative of the pristine marine environment in the winter and strongly biased the correlations between BC and CO, sulfate, and organics. Therefore, we removed the affected datapoints from November 12th and 14th, 2015 spirals and re-evaluated the correlations abovementioned. Next based on the new results, we have corrected the discussion in section 3.2 as following (**Page 7, Line 17** of the track changes revised version of the manuscript):

~~No correlations between CO and BC, BC and sulfate, and BC and organics are found during NAAMES 1 (the linear regression R^2 values are 0.04, 0.25, and 0.20, respectively) (Fig. 6a, b, and c) indicating a predominantly low aerosol environment with minor influence of anthropogenic pollution and ocean emissions.~~ No correlations between CO and BC, BC and sulfate, and BC and organics are found in the MBL (Fig. 7a, b, and c). Similarly, despite higher level of BC in the FT, we could not find correlations between FT BC and FT CO (linear regression R^2 values is 0.01) (Fig. 7d). On the other hand, the correlations between BC and sulfate, and BC and organics in the FT is moderate (the linear regression R^2 values are 0.64, and 0.40, respectively) (Fig. 7e, and f). These results indicate that, although the predominantly low aerosol environment with only reduced ocean emissions, the WNAO in the winter is subjected to minor episodes of anthropogenic pollution, such as ship-traffic in the MBL and transport of continental polluted air masses in the FT) that influence the atmospheric vertical column.

Fig. 6a, b, and c, in the revised manuscript **Fig. 7a, b, c, d, e, f** have been modified and are copied below.

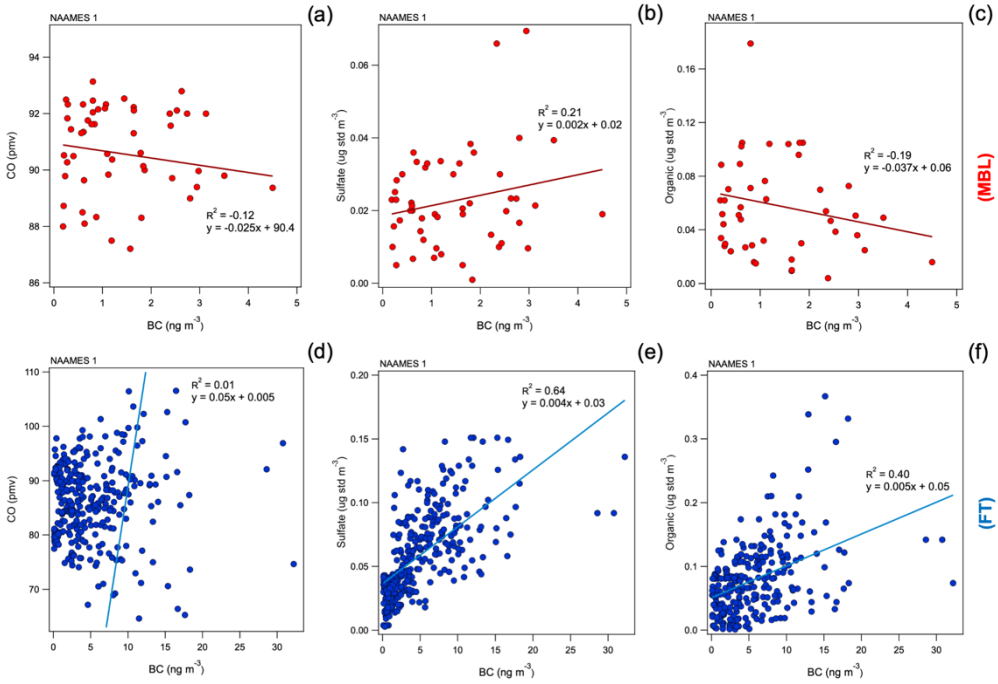


Figure 6. Correlation between BC and CO (a, d), BC and sulfate (b, e), and BC and organics (c, f) in the MBL (red) and FT (blue) during NAAMES-1

3. Minor corrections: *page 10, line 24: should be "3.3.2"; line 27: duplicate "%". ?*

Response: We thank Referee #1 for noticing this discrepancy in the text, "3.3.1 CCN seasonal variations" is incorrect and the number of the section has been changed as following (**Page 12, Line 10** of the track changes revised version of the manuscript):

"3.3.2 CCN seasonal variations"

The duplicate "%" page 10, line 27 has been removed.

Responses to Anonymous Referee #2

Comment: *This study presents and discusses a compilation of datasets of airborne aerosol measurements performed during three observation programs in the Western North Atlantic Ocean (WNAO). Aircraft campaigns provide unique information about the three-dimensional structure of the atmosphere. However, they typically provide only "snapshots" of the atmospheric composition in a given area. Here the Authors make an attempt to overcome this limitation by combining the datasets from 37 spirals performed in a similar altitude range and by lumping them into three sets representative of different seasons: winter, spring and summer. The results provide a clear picture of several relevant patterns in aerosol properties and chemical tracers in the marine boundary layer and in the free troposphere in the WNAO, as well as information about the processes driving the variability of such parameters as a function of the seasonal variations in the meteorological conditions, biological activity in surface ocean and atmospheric dynamics. The discussion is clear and concise, supported by good graphics. The paper misses a table summarizing the main statistics of meteorological variables and of the concentrations of carbon monoxide, DMS and aerosol parameters in the MBL and FT in the three seasons. A figure with the average vertical profiles of the main aerosol parameters in the three seasons would of help, too. Having said that, in this reviewer's opinion, describing the great variability in the individual vertical profiles just focusing on the seasonal difference is an oversimplification: Fig. 6 (especially panels d, e, g, h) suggests that very different events with contrasting compositions were lumped together. It would be important, therefore, to take the correlations in Fig. 6 with a grain of salt, because in several cases the datasets do not look homogeneous at all. At the same time, it would be important to add a sentence in the Abstract to acknowledge the intra-seasonal variability.*

Response: We thank Anonymous Referee #2 for the constructive criticism and suggestions. Per the suggestions and agree with Referee #2, we added **Table 3 (page 25)** which provide a summary statistic of meteorological parameters, trace gasses and aerosol properties during the three NAAMES field campaigns**, Fig. 3** (**page 28**) showing seasonal averaged vertical profiles of BC, CO, sulfate, organic, N_{CN}, and N_{CCN}, reviewed Fig. 6 **(Fig. 7, 8 , and 9 (page 33-34)** in the revised version of the manuscript), improved Sect. 3.2 discussion (**page 7, line 40**), and added a sentence in the Abstract to highlight the importance of the intra-seasonal variability observed (**page 1, line 34**). We too agree that the revised manuscript is overall stronger, and capable of higher impact. All detailed comments are addressed in the following point-by-point discussions below. All the alterations to the manuscript are show in the track changes revised version of the manuscript attached to this document - the original Referee #2 comments are highlighted in italic black and our responses follow in blue. (blue).

- A table providing a summary statistic of meteorological parameters, trace gasses and aerosol properties during the three NAAMES field campaigns has been added to the track changes revised version of the manuscript as **Table 3 (page 25)**

Table 3. Summary statistic of meteorological parameters, trace gasses, and aerosol properties measured during the three NAAMES field campaigns. The numbers are shown as mean ± standard deviation. N_{CN} and N_{CCN} values indicate geometric mean ⁎ geometric standard deviation.
* MBL BC levels removing data affected by the ship-traffic events discussed in Section 3.2.

Parameters	NAAMES-1	NAAMES-2	NAAMES-3
MBL Height	1776 ± 313	1657 ± 445	1144 ± 311

	MBL	FT	MBL	FT	MBL	FT
θ (K)	279 ± 8	298 ± 8	296 ± 11	302 ± 9	292 ± 5	315 ± 9
***W* (ppmv)**	5204 ± 2937	2001 ± 2712	5422 ± 4404	3475 ± 2640	8323 ± 1928	2008 ± 2201
RH (%)	78 ± 15	43 ± 26	55 ± 27	45 ± 23	63 ± 19	22 ± 17
CO (ppmv)	90 ± 3.3	86 ± 7.2	104 ± 8.5	109 ± 18.8	112 ± 17	107 ± 32.7
BC (ng m^{-3})	8.3 ± 21 *(1.8 ± 0.9)	5.1 ± 4.8	9.4 ± 8	30.7 ± 35	21 ± 28	15 ± 28
DMS			0.034 ± 0.03		0.014 ± 0.02	
Sulfate (ug std m^{-3})	0.03 ± 0.03	0.07 ± 0.03	0.2 ± 0.16	0.18 ± 0.11	0.25 ± 0.16	0.14 ± 0.07
Organic (ug std m^{-3})	0.07 ± 0.03	0.08 ± 0.06	0.25 ± 0.18	0.77 ± 1.21	1.03 ± 1.12	1.03 ± 1.12
N_{CN} (cm^{-3})	96 ∗ 2.6	286 ∗ 1.9	345 ∗ 2	553 ∗ 2	442 ∗ 1.9	860 ∗ 1.6
N_{CCN} (cm^{-3})	15 ∗ 2.3	41 ∗ 2.4	72 ∗ 2.2	89 ∗ 3.3	79 ∗ 3.5	61 ∗ 3.4

- A figure showing seasonal averaged vertical profiles of BC, CO, sulfate, organic, N_{CN}, and N_{CCN} has been added as **Fig. 3 (page 28)** to the track changes revised version of the manuscript:

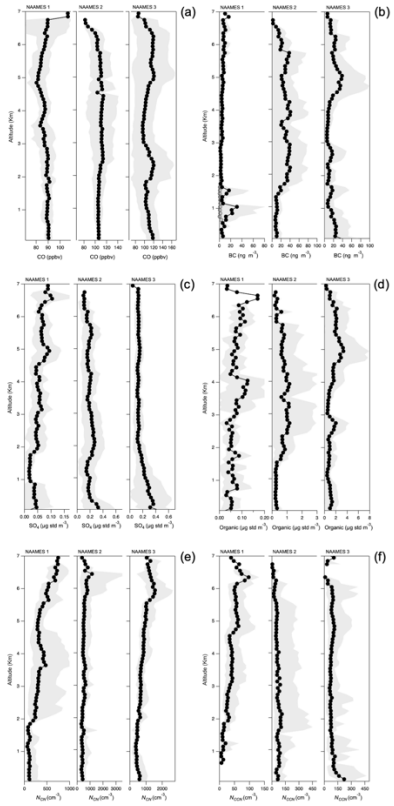
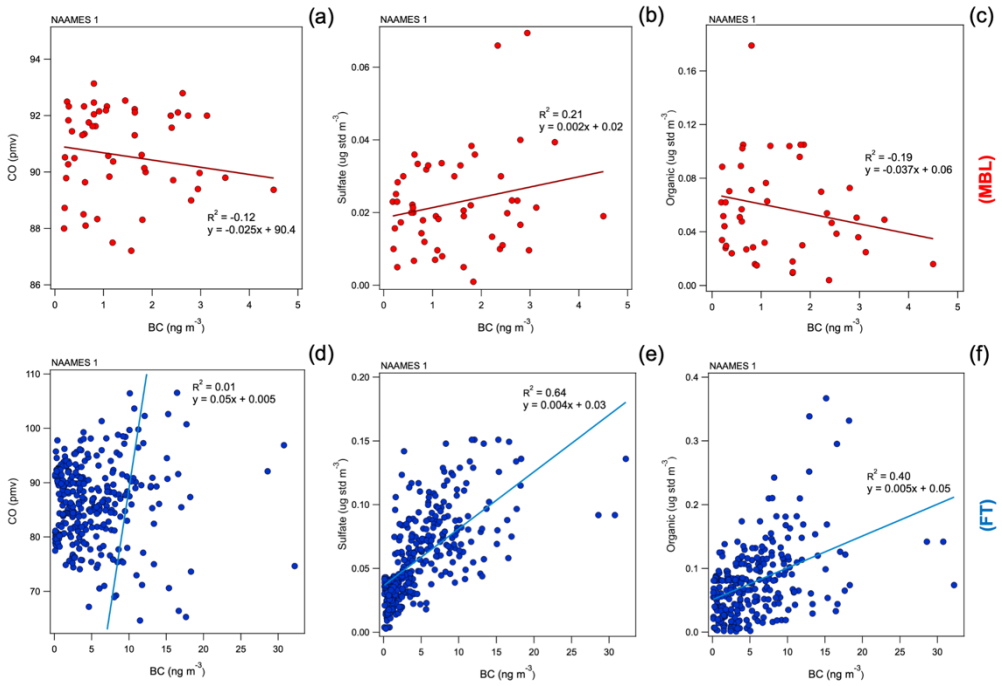
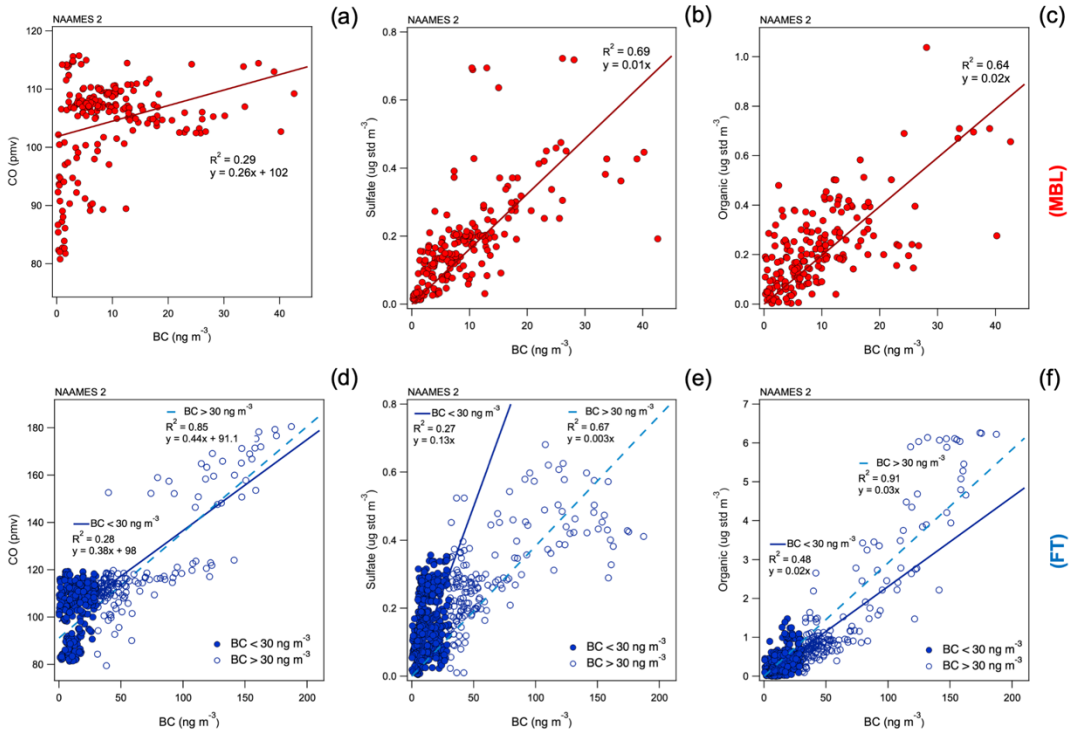


Figure 3. Vertical profiles of mean and standard deviation (grey shadow) of CO (a), BC (b), sulfate (c), organic (d), and geometric mean ∗ geometric standard deviation of N_{CN} (e), and N_{CCN} (f) during the three NAAMES field campaigns. Empty grey dots in Fig. b, represent mean of BC when data affected by the ship-traffic events discussed in Section 3.2 are removed.

- Fig. 6 has been reviewed and split in three different figures: **Fig. 7, 8, and 9** (page 33-34) of the track changes revised version of the manuscript:



- Figure 7. Correlation between BC and CO (a, d), BC and sulfate (b, e), and BC and organics (c, f) in the MBL (red) and FT (blue) during NAAMES-1.



- Figure 8. Correlation between BC and CO (a, d), BC and sulfate (b, e), and BC and organics (c, f) in the MBL (red) and FT (blue) during NAAMES-2.

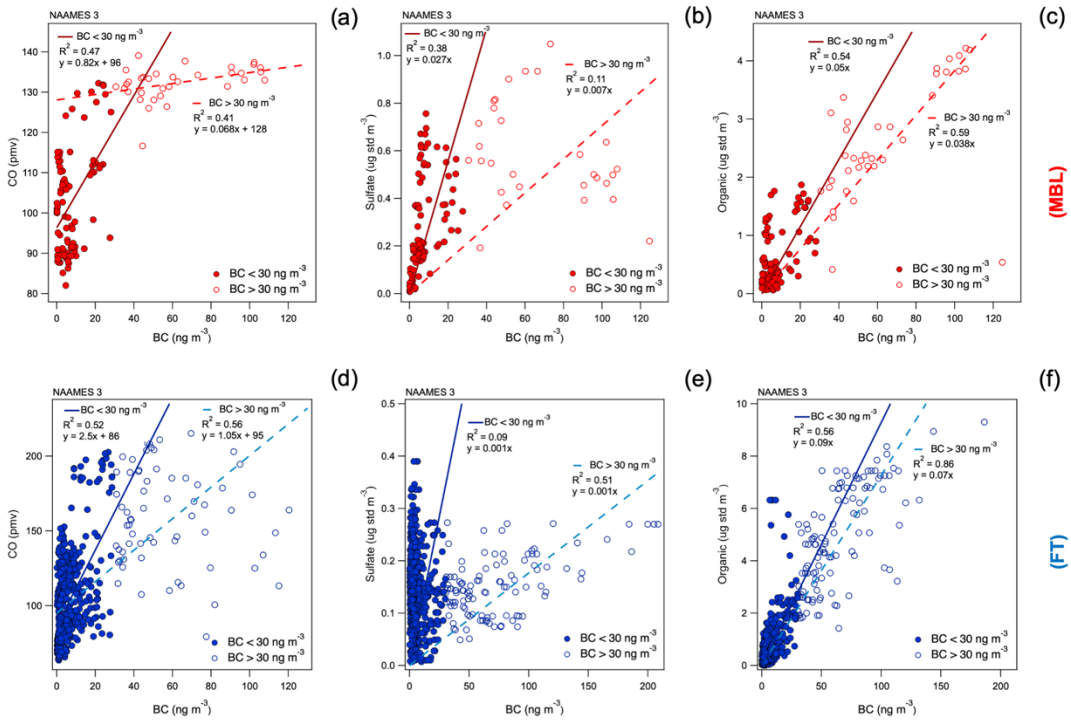


Figure 9. Correlation between BC and CO (a, d), BC and sulfate (b, e), and BC and organics (c, f) in the MBL (red) and FT (blue) during NAAMES-3.

- The importance of the intra-seasonal variability observed has been added to the abstract (**page 1, line 34** of the track changes revised version of the manuscript):

> The vertical distribution of submicron aerosol particles exhibited strong seasonal variation, as well as elevated intra-seasonal variability depending on emission sources and aerosol processes in the atmospheric column.

Specific Comments:

Section 3.2 NAAMES-1. *Fig. 6b shows that most of the data belong to a cluster showing a positive correlation between sulfate and BC, at least for sulfate concentrations greater than 0.05 ug m-3. The three flights performed on 11/12 and 11/14 are characterized by FT levels of BC and sulfate somewhat higher than the background.*

Response: Per the suggestion of Anonymous Referee #1, we have constrained the origin of elevated BC concentrations in the MBL on November 12th and 14th, 2015 (Page. 6, line 40 of the revised manuscript). We found that the enhanced BC levels in the MBL in the days above-mentioned are likely related to ship-traffic emissions. On recommendation of Referee #1 we excluded the BC measurements affected which are not representative of the pristine marine environment in the winter and re-assessed the correlation between BC and CO, sulfate, and organics. The revised correlations are shown in Fig. 6d, e, and f, and discussed in section 3.2 (**Page. 6, line 40** of the track changes revised version of the manuscript).

> During NAAMES-1, we observe a mean CO concentration = 87.2 ± 6.6 ppbv and no significant variations within days. CO vertical profiles show similar concentrations at all the altitudes sampled, being only

slightly higher in the MBL (mean = 90.1 ± 3.3 ppbv) than in the FT (mean = 86.1 ± 7.2 ppbv) ~~(Fig. 3a, Fig. SI.1.a, and Fig. SI.2.a)~~ (Table 3, Fig. 3a, Fig. 4a, and Fig. SI.1.a). BC concentrations are also generally low ~~(mean of 6.2 ± 11.2 ng m^{-3})~~ ~~(mean of 8.3 ± 21 ng m^{-3} in the MBL, and 5.1 ± 4.8 ng m^{-3} in the FT)~~, characteristic of unpolluted conditions observed during the winter (Table 3, Fig. 3b, Fig. 4b, and Fig. 6a). The only exceptions are Nov. 12th, 2015 and Nov. 14th, 2015 when BC exhibits peaks in the MBL up to 101.4 ng m^{-3} and 106.2 ng m^{-3} respectively, at altitudes < 1500 m ~~(Fig. 3b, and Fig. SI.2.a.1-3)~~ (Fig. 4b, and Fig. SI.2.a.1-3). The enhanced BC levels might be the result of ship-traffic emissions. Analysis of 10-days Flexapart backtrajectory indicate the arrival of air masses from the Artic suggesting clean maritime influence. Furthermore, Worlview FIRMS VIIRS Fire and Thermal Anomalies products show absence of wildfires on the air mass transport path prior the arrival to the NAAMES-1 domain, therefore excluding the contributions from biomass burning to the BC level observed. After removing the BC measurements affected by ship-traffic emissions (data discarded altitude < 1500 m on Nov. 12th, 2015, and altitude < 1050 m on Nov. 14th, 2015) we found mean BC concentrations in the MBL of 1.8 ± 0.9 ng m^{-3} and lower than in the FT. ~~the intrusion of polluted air masses from continental origins due to wintertime low-altitude long-range transport. Field studies conducted in several locations over the North Atlantic ocean region have shown background concentrations of BC ranging between 10 and 40 ng m^{-3} under unperturbed marine conditions (O'Dowd et al., 2004; Shank et al., 2012; Pohl et al., 2014; Cavalli et al., 2016), while BC concentration up to 600 ng m^{-3} have been reported during periods of time affected by intense continental emission plumes (Corrigan et al., 2008).~~ Non-refractory aerosol chemical composition analysis also reveals low concentrations of sulfate (0.05 ± 0.03, 0.03 ± 0.03, and 0.07 ± 0.03 µg std m^{-3} respectively at the surface ocean, in the MBL, and in the FT) ~~(Fig. 4a, Fig. 5b, and Fig. SI.2.a)~~ (Fig. 3c, Fig. 5a, and Fig. 6b), and organic (0.1 ± 0.07, 0.05 ± 0.08, and 0.08 ± 0.06 at the surface ocean, in the MBL, and in the FT, respectively) aerosol mass ~~(Fig. 4b, and Fig. SI.1.b, and Fig. SI.2.a)~~. ~~No correlations between CO and BC, BC and sulfate, and BC and organics are found during NAAMES-1 (the linear regression R^2 values are 0.04, 0.25, and 0.20, respectively) (Fig. 6a, b, and c) indicating a predominantly low aerosol environment with minor influence of anthropogenic pollution and ocean emissions.~~ No correlations between CO and BC, BC and sulfate, and BC and organics are found in the MBL (Fig. 7a, b, and c). Similarly, despite higher level of BC in the FT, we could not find correlations between FT BC and FT CO (linear regression R^2 values is 0.01) (Fig. 7d). On the other hand, the correlations between BC and sulfate, and BC and organics in the FT is moderate (the linear regression R^2 values are 0.64, and 0.40, respectively) (Fig. 7e, and f). These results indicate that, although the predominantly low aerosol environment with only reduced ocean emissions, the WNAO in the winter is subjected to minor episodes of anthropogenic pollution, such as ship-traffic in the MBL and transport of continental polluted air masses in the FT) that influence the atmospheric vertical column.

Section 3.2 NAAMES-2. *There is a lot of variability between days. The flight of 5/20 is characterized by very high FT concentrations of carbonaceous aerosols. By contrast, on 5/26, the air column was depleted of carbonaceous aerosols while sulfate exhibited moderate concentrations in the FT. The following days showed relatively high concentrations of all aerosol compounds in the lower FT. The discussion does not acknowledge such intra-seasonal variability, nor it takes into account possible source regions for the air masses.*
Section 3.2 NAAMES-3. *The compositions observed in this period encompass at least three different conditions. The main difference is between the profiles recorded on 09/08 and 09/08 which were characterized by low carbonaceous aerosol concentrations and high sulfate concentrations and the days before (09/04 and 09/06) or the following ones (09/12 to 09/17) when a multi-layer structure was observed.*

Response: During NAAMES-2 and -3, the C-130 aircraft intercepted aerosol plumes with different origins which affected the regional pristine marine environment and led to high inter-day variability. While the investigation of specific single events is beyond the scope of this study and will be subject of further works, we do agree that Section 3.2 is not complete without further explanations of the variability observed between flights in spring and summer. We have improved the discussion and incorporated information to clarify the presence of non-uniform datasets within flights in the revised manuscript as follow (**Page. 7, line 25** of the track changes revised version of the manuscript):

> The highest mean CO and BC values are observed during NAAMES-2, being respectively 107.9 ± 15.7 ppbv and 24.8 ± 32.1 ng m^{-3}. Maximum CO and BC concentrations in the springtime have been previously reported over the North Atlantic and are related to more frequent contributions from North American continental outflow which is lofted into the FT and transported over the ocean by westerly frontal passages and convection that occasionally mix emissions down into the MBL (Zhao et al., 2012; Wood et al., 2015; Zheng et al., 2018). Namely, field studies conducted in several locations over the North Atlantic ocean region have shown background concentrations of BC ranging between 10 and 40 ng m^{-3} under unperturbed marine conditions (O'Dowd et al., 2004; Shank et al., 2012; Pohl et al., 2014; Cavalli et al., 2016), while BC concentration up to 600 ng m^{-3} have been reported during periods of time affected by intense continental emission plumes (Corrigan et al., 2008). Along the vertical column, mean CO levels remain almost constant (mean MBL CO = 104.4 ± 8.5 ppbv, and mean FT CO = 109 ± 18.8 ppbv) ~~(Fig. 3a, Fig. SI.1.a, and Fig. SI.2.b)~~) (Table 3, Fig. 3a, Fig. 4a, and Fig. SI.1.a), while in the case of BC, we observe a well-defined vertical trend with 3-fold lower concentrations in the MBL (mean MBL BC = 9.4 ± 8 ng m^{-3}) than in the FT (mean FT BC = 30.7 ± 35 ng m^{-3}) ~~(Fig. 3b, Fig. 4a, and Fig. SI.2.b).~~ (Table 3, Fig. 3b, Fig. 4b, and Fig. 6a). It is important noting that BC concentrations in the FT also have a statistically relevant higher variability due to the occurrence of high-concentration BC events (BC > 30 ng m^{-3}) during some of the flights analyzed, namely on May 20th, 28th, 29th and 30th, 2016 (Fig. 3b). Excluding the above-mentioned events, BC concentrations are still higher than what observed during NAAMES-1, suggesting minor but persistent contributions from continental polluted emissions over the entire period. Overall, BC is not well correlated with CO (linear regression R^2 = 0.29, and R^2 = 0.28, respectively in the MBL and FT) but the correlation improves in the FT when BC concentrations are > 30 ng m^{-3} (linear regression R^2 = 0.79) (Fig.8a, and d). ~~MBL BC is not well correlated with CO (linear regression R^2 = 0.29), but the correlation improves in the FT (linear regression R^2 = 0.79) (Fig. 6d).~~ Possible explanations for this are different CO and BC lifetimes and removal mechanisms ~~in the MBL and FT~~. Indeed, while CO has an atmospheric lifetime of months and its concentration is mainly driven by hydroxyl radical (OH) oxidation (Novelli et

al., 1998; Seinfeld and Pandis, 2016), BC lifetime is shorter and it is removed through cloud and precipitation scavenging processes (Cape et al., 2012; Bond et al., 2013). Sulfate loadings are are 0.2 ± 0.08 μg std m^{-3} at the surface ocean, 0.2 ± 0.16μg std m^{-3} in the MBL, and 0.18 ± 0.11 in the FT,(Table 3, Fig. 3c, Fig. 5a, and Fig. 6b), and the mixing ratio of DMS in the MBL during NAAMES-2 d the highest id the highest observed during the three field campaign (values up to 0.325 ppbv) consistent with other works conducted in the North Atlantic region (Yoon et al., 2007; Quinn et al., 2019; Saliba et al., 2020; Sanchez et al., 2021) (Fig. 4c). The organic mean loadings and associated standard deviations follow a different trend being lower in the MBL (0.4 ± 0.15 μg std m^{-3} at the surface ocean, and 0.25 ± 0.18 μg std m^{-3} in the MBL) and more elevated in the FT (0.77 ± 1.21 μg std m^{-3}) (Table 3, Fig. 3d, Fig. 5b, Fig. SI.1.b). In the MBL the correlation between BC and sulfate is moderate (R^2 = 0.69) suggesting that part of the sulfate has marine origin (Fig. 7b). Simultaneously, in the FT we found linear regression R^2 = 0.27 when BC concentrations are < 30 ng m^{-3} and R^2 = 0.67 and for BC > 30 ng m^{-3}(Fig. 7f). The weak correlation between sulfate and BC when BC is < 30 ng m^{-3} is driven by the measurements collected on May 19th and 26th, 2016. Excluding those spirals, the linear regression would generate an R^2 = 0.61, in agreement to what observed in the MBL and in the FT for BC > 30 ng m^{-3}. This result might indicate sulfate mass production from DMS oxidation in the FT occurring after the loft of DMS from the MBL. Similar results were observed for the correlation between BC and organic being the linear regression R^2 values 0.64 in the MBL, and 0.42 and 0.91 in the FT respectively for BC < 30 ng m^{-3} and BC > 30 ng m^{-3}. Again, the modest correlation found in the MBL provide evidence of an additional source of organic particles not related to long-range transport of continental air masses. In a recent field study conducted in the Arctic, Mungall et al. (2017) showed that the sea surface microlayer at the ocean-atmosphere interface is enriched by dissolved organic carbon content (e.g. surfactants and plankton exudates) and can be an important source of oxygenated volatile organic compounds in the MBL which can be lofted to the FT and , possibly contribute to the total sub-micron non-refractory mass along the vertical column. Simultaneously, in the FT, the stronger correlation between BC and organic when BC > 30 ng m^{-3} is accompanied by mean organic mass ~4-fold higher during periods when BC < 30 ng m^{-3} indicating the contributions from anthropogenic sources of continental origins with high organic components. ~~Sulfate and organic mean loadings are respectively 0.2 ± 0.08, and 0.4 ± 0.15 μg std m^{-3} at the surface ocean, 0.2 ± 0.16, and 0.25 ± 0.18 μg std m^{-3} in the MBL, and 0.18 ± 0.11, and 0.77 ± 1.21 μg std m^{-3} respectively in the FT (Fig. 4a, and Fig. 5b – sulfate, Fig. 4b, and Fig. SI.1.b – organic, and Fig. SI.2.b). The correlation between BC and sulfate is moderate (R^2 = 0.69) and similar in MBL and FT (Fig. 6e), while a stronger correlation is observed between BC and organics (R^2 = 0.91) (Fig. 6f), especially in the FT. In accordance with the results of (Sanchez et al., 2018), the above results indicate contributions from anthropogenic sources of continental origins, especially in the case of the organic components for which we observed mean organic mass ~4-fold higher when BC concentrations > 40 ng m^{-3}. However, the lower correlation between BC and sulfate also suggest that part of the sulfate has marine origin. Similarly, if considering only the MBL, the R^2 obtained comparing BC and organics is lower (0.64), suggesting an additional source not related to long range transport of particles. In a recent field study conducted in the Arctic, Mungall et al. (2017) showed that the sea surface microlayer at the ocean-atmosphere interface is enriched by dissolved organic carbon content (e.g.~~

~~surfactants and plankton exudates) and can be an important source of oxygenated volatile organic compounds in the MBL, possibly contributing to the total sub-micron non-refractory mass.During the NAAMES-2 field campaign, we find the highest mixing ratio of DMS in the MBL (values up to 0.325 ppbv) consistent with other works conducted in the North Atlantic region (Yoon et al., 2007; Quinn et al., 2019; Saliba et al., 2020; Sanchez et al., 2021) (Fig. 3c, and Fig. SI.2.b).~~

During NAAMES-3, CO and BC concentrations vary significantly within flights ~~(Fig. 3a, Fig. SI.1.a - CO, and Fig. 3b, Fig. 4a - BC)~~ (Fig. 3a, Fig. 4a, Fig. SI.1.a - CO, and Fig. 3b, Fig. 4b, Fig. 6a - BC). The lowest mean CO and BC concentrations are observed on Sept. 08th and 09th (CO mean of 83.5 ± 6.5 ppbv, BC mean 3.9 ± 3.4 ng m^{-3}) and remain relatively constant throughout the vertical column (Fig. SI.2.c.27-31). Minimum CO levels in the summer have been reported by several studies (Honrath, 2004; Wood et al., 2015; Zheng et al., 2018) and are found to be associated with photolytic destruction due to high summertime OH levels (Novelli et al., 1998). However, the analysis conducted on the data collected on the other days of the NAAMES-3 field campaign show higher mean and standard deviation CO and BC values of respectively 124.7 ± 27.3 ppbv and 22.3 ± 32.5 ng m^{-3}. High concentrations of CO and BC have been related to the long-range transport of continental biomass burning plumes over the WNAO due to the North American and Canadian wildfire season (Honrath et al., 2004; Val Martín et al., 2006). ~~CO of 124.7 ± 27.3 ppbv and BC of 22.3 ± 32.5 ng m^{-3} and a non-uniform distribution along the vertical columns characterized by ~ 800 to 2000 m height layers with CO levels > 200 ppbv and BC concentrations > 100 ng m^{-3}.~~ Low-level altitude (below ~3000 m) transport of CO from North America to the Atlantic Ocean has been observed in summer by earlier studies (Li et al., 2005; Owen et al., 2006), while Zheng et al. (2020) reported upper troposphere transport of biomass burning particles from western Canadian wildfire over the Eastern North Atlantic in late August 2017. The distributions of CO and BC along the vertical columns are non-uniform characterized by ~ 800 to 2000 m height layers with CO levels > 200 ppbv and BC concentrations > 100 ng m^{-3} at different altitudes depending on the day, or even on the time of the day. Here we observe the highest concentration of CO and BC and diverse multi-layer structures on September 4th, 6th, and 12th 2017. Namely, on Sept. 4th we found CO > 200 ppmv and BC > 30 ng m^{-3} at altitudes > 4.2 Km and in the residual layer (1. 2 to 1.8 Km), which entrained into the MBL in the late afternoon (spiral 2 at 16:07 UTC). Similar enhanced CO and BC concentrations were also observed on Sept 6th between 1.9 and 3 Km and in the MBL, and on Sept. 12th in the FT at altitudes > 4 Km (Fig. 4a, and b). ~~and high concentrations of CO and BC have been related to the long-range transport of continental biomass burning plumes over the WNAO due to the North American and Canadian wildfire season (Honrath et al., 2004; Val Martín et al., 2006).The linear regression obtained comparing BC and CO generate an R^2 = 0.54 (Fig. 6g).~~ Correlation between BC and CO is moderate and slightly lower in the MBL (R^2 = 0.47 for BC < 30 ng m^{-3} and R^2 = 0.41 for BC > 30 ng m^{-3}) than in the FT (R^2 = 0.52 for BC < 30 ng m^{-3} and R^2 = 0.57 for BC > 30 ng m^{-3})(Fig. 9a, and d). Our result is consistent with Val Martín et al. (2006) who observed higher variability in BC than CO at Pico Mountain (in the Eastern North Atlantic) due to different fire emission rates and wet scavenging during the transport to the site. Mean mass concentrations of non-refractory sulfate are 0.12 ± 0.13 µg std m^{-3} at the surface ocean, 0.25 ± 0.16 µg std m^{-3} in the MBL, and 0.14 ± 0.07 µg std m^{-3} in the

FT (Table 3, Fig. 3c, Fig. 5a, and Fig. 6b). Compared to NAAMES-2 sulfate mass in the MBL is higher, and mixing ratio of DMS is lower than in the spring being up to 0.11 ppbv in the MBL (Fig. 4c). These results are likely related to the combination of of phytoplankton abundance and productivity reduction at the end of the summer / beginning of fall which cause DMS levels to decline (Lana et al., 2011), and increased sunlight and enhanced concentrations of OH which lead to DMS oxidation sulfate mass production (Zawadowicz et al., 2020). .. Supporting our hypothesis, highest concentrations of sulfate in the MBL are associated to the lowest level of DMS on Sept. 6th, 8th, and 9th, while we observed the opposite trend on Sept. 4th, 16th, and 17th, 2017 (Fig. 4c, and Fig. 5a). In the FT, sulfate concentrations are lower than during NAAMES-2. Accordingly, the correlation between BC and sulfate is weak and only slightly improves in the FT when BC concentration are > 30 ng m^{-3} (R^2 = 0.38 for BC < 30 ng m^{-3} and R^2 = 0.11 for BC > 30 ng m^{-3} in the MBL, and R^2 = 0.09 for BC < 30 ng m^{-3} and R^2 = 0.51 for BC > 30 ng m^{-3} in the FT) (Fig. bc, and e), and excluding the presence of a strong anthropogenic source of sulfate. Mean mass concentrations of non-refractory organic are 0.49 ± 0.42 µg std m^{-3} at the surface ocean, 1.03 ± 1.12 µg std m^{-3} in the MBL, and 1.03 ± 1.12 µg std m^{-3} respectively in the FT (Table 3, Fig. 3d, Fig. 5b and Fig. SI.2.b), and higher than during the other two NAAMES field campaigns. The correlation between BC and organic is moderate and R^2 ~ 0.5 in both MBL and FT (R^2 = 0.54 for BC < 30 ng m^{-3} and R^2 = 0.59 for BC > 30 ng m^{-3} in the MBL, and R^2 = 0.56 for BC < 30 ng m^{-3} in the FT), (Fig. 9c, and f), however when BC > 30 ng m^{-3} and mean organic mass are ~6-fold higher and the linear regression between BC and organic generate an R^2 = 0.5. for. Our observations are supported by ~~and organic are, respectively, 0.12 ± 0.13 and 0.49 ± 0.42 µg std m^{-3} at the surface ocean, 0.25 ± 0.16, and 1.03 ± 1.12 µg std m^{-3} in the MBL, and 0.14 ± 0.07, and 1.03 ± 1.12 µg std m^{-3} respectively in the FT (Fig. 4a, and Fig. 5b - sulfate, Fig. 4b, and Fig. SI.1.b - organic, and Fig. SI.2.c). No correlation is found between BC and sulfate (R^2 = 0.21) (Fig. 6h), and sulfate concentrations in the FT are lower than during NAAMES 2 therefore excluding the presence of a dominant anthropogenic source of sulfate. Instead, enhanced sulfate mass concentrations over the Atlantic Ocean in summertime have been previously attributed to DMS production and high oxidant levels at the surface ocean (Zawadowicz et al., 2020). Conversely, organic aerosol loadings are higher than what was observed during the other two NAAMES field campaigns. The correlation between BC and organic have R^2 = 0.89 (Fig. 6i), and mean organic mass were 6-fold higher when BC concentrations were > 40 ng m^{-3}. P~~previous work has shown that in biomass burning plumes, aerosol composition is typically dominated by organics which can cover BC particles with a thick coating (Ditas et al., 2018), while concentrations of sulfate are lower and increase with aging (Schlosser et al., 2017). ~~During NAAMES 3, mixing ratio of DMS were lower than in the spring being up to 0.11 ppbv in the MBL (Fig. 3d) likely due to the decline of phytoplankton abundance and reduced productivity at the end of the summer / beginning of fall (Lana et al., 2011). Increased sunlight in summertime may also have caused quicker DMS oxidation than in spring led by enhanced concentrations of OH.~~

Fig. 1c. *Do the three flights done on 05/26 exactly share the same RH profile?*

Response: Relative humidity measurements from the meteorological sensor package are not available for the spirals conducted on May 26th, 2016. To overcome this limitation and still be able to characterize the vertical structure of the atmospheric column, we used the RH measurement collected via radiosonde ship-launch on May 26th, 2016 at 2:26pm UTC time. We thank Referee #2 for pointing out the lack of this information in our manuscript. We have incorporated this information in Fig. 1 caption on **Page 26, line 2** of the track changes revised version of the manuscript.

> **Figure 1.** Vertical profiles potential temperature (a), H2O mixing ratio (b), and relative humidity (c) during NAAMES-1, -2, and -3. Relative humidity measurements from the meteorological sensor package are not available for the spirals conducted on May 26th, 2016, therefore RH measurement collected via radiosonde ship-launch on May 26th, 2016 at 2:26pm UTC time.

Fig. 1c. *The units for BC should be ng m-3 not ug m-3.*

Response: We thank Referee #2 for catching the typo in our Fig. 3b label axes. The label axes have been corrected to "ng m^{-3}" as shown below.