

We thank the reviewer for their comments. Below we reproduce the comments in blue and provide our discussion in black.

The authors seem to have spent very little effort seeking prior data about aerosol chemistry projects near their study area, even though Azores and surrounding ocean have been a pretty common site for field projects. They did not have the time resolution, CIMS or CCN, but the sulfur chemistry in the central North Atlantic has been studied at least as far back as 1987 (Galloway, Penner, et al. 1992; Galloway, Keene, et al. 1990; Pszenny et al. 1990) and the ASTEX/MAGE project, employing aircraft, ground stations, and a ship, was based in the Azores in 1992 (Blomquist, Bandy, and Thornton 1996; Huebert et al. 1996; Zhuang and Huebert 1996). I was really puzzled that no mention was made of them. The ACE-2 project (Quinn et al. 2000; Raes et al. 2000) was centered a bit farther south, but seems relevant, as do some oceanographic cruises (e.g. Andreae et al. 2003).

We are now briefly summarizing these previous North Atlantic measurements in the Introduction, “The atmospheric chemistry of North Atlantic region, including the Azores, has been studied in previous oceanographic cruises (Andreae et al., 2003) and aircraft campaigns. Early work on sulfur cycle in the Eastern North Atlantic has been summarized by Galloway et al. (1992). Notable campaigns in the region include the Atlantic Stratocumulus Transition Experiment/Marine Aerosol and Gas Exchange (ASTEX/MAGE) (Blomquist et al., 1996) and the second Aerosol Characterization Experiment (ACE-2) (Raes et al., 2000).”

Some of these papers are also referenced later in the discussion of MSA:SO<sub>4</sub> ratios, “The MSA to non-sea salt sulfate ratio (MSA:SO<sub>4</sub>) measured during ACE-ENA in the summer was 0.02 on average in the MBL (<1000 m), which is lower than historical estimates of the ratio. For example, Pszenny et al. (1990) reports the ratio as 0.05 in the North Atlantic in August-September, Berresheim et al. (1991) reports 0.033 in Western North Atlantic in September, Savoie et al. (2002) reports 0.06 in Bermuda in September and 0.05 in Mace Head in August. Huebert et al. (1996) found 0.07 in marine air masses and 0.02 in continental air masses in June in the Azores. The measurements reported in those earlier studies are based on analysis of filter samples, however, and they may not be directly comparable to AMS measurements reported here.”

The authors apparently made the rather peculiar decision that only other projects featuring AMS measurements were worth considering, resulting in rather tenuously related comparisons (the Amazon basin, and biomass burning plumes from Southern Africa, for example.) Even then, there are more appropriate comparisons than the ones used. VOCALS looked at another clean cloud deck (Shank et al. 2012; Wood et al. 2011). ATOM passed through your study area 4 times with a huge payload, including AMS and CIMS (Wofsy et al. 2018).

The comparison with the Amazon basin was made to highlight the cleanliness of the ENA MBL measured in this campaign. The other comparisons made in section 3.1.1 on p. 7 are made on the basis of similarity of marine conditions (Polarstern and NAAMES cruises) and geography (NEAQS flights that occurred on the East coast of the U.S. which would be a source region for aged emissions seen at ENA). We have now added some non-AMS comparisons, cited in the previous reply also centering on the East Atlantic region. We have decided not to compare with

ATom and VOCALS explicitly in order not to clutter section 3.1.1, especially since the location for VOCALS was the Pacific Ocean off the coast of South America. The ATom campaign is also discussed in the literature review portion of the introduction, “Between 2016 and 2017, AMS was also deployed aboard the NASA DC-8 aircraft during the Atmospheric Tomography (ATom) missions in the remote atmosphere, including the North Atlantic region (Hodshire et al., 2019).”

I cannot fathom why there is no mention of any of the many studies based at Mace Head, on the west coast of Ireland (e.g. Dall’Osto et al. 2010; Ovadnevaite et al. 2014).

We added the following, “Of note are also studies on the west coast of Ireland at the Mace Head observatory, which frequently encounter North Atlantic air masses, as described in Dall’Osto et al. (2010) and Ovadnevaite et al. (2014).”

The meteorological setting is woefully incomplete. What was typical cloud height? Was there usually an extensive stratocumulus deck? What altitude? Was the boundary layer decoupled? Was there a strong inversion? Was there a buffer layer? Was there drizzle that might be scavenging aerosol? Why were the altitude ranges 0 m to 1000 m and 1000 m to 3000 m chosen? I presume it had to do with boundary layer depth, but that should be explicit. Using fixed altitude ranges is convenient, but does it really reflect the structure of the atmosphere? It is often best to group data into mixed layer, cloud layer, buffer layer (if present), and free troposphere.

This information has been included in more detail in an overview of the ACE-ENA campaign submitted to the Bulletin of American Meteorological Society (Wang et al., 2021). Repeating it in this paper is out of its scope, as it is not intended to be a comprehensive review of the campaign.

Are there estimates of entrainment velocity during the experiment? That would constrain how fast the MBL is diluted with FT air and thus give a lower bound on MBL sources of aerosol. Or was the meteorological situation too complex for such approximations to make sense?

To our knowledge such estimates have not been made to date.

Since the literature survey was so minimal, I suppose it was inevitable that there would be a lack of direct comparison with earlier data from the region. But it would be interesting to know whether things have changed. Of particular interest is the fact that earlier experiments found much more  $MS^-$ . Is that a real change, or is it possibly that much of the  $MS^-$  was on particles too large for the AMS? Were the  $MS^-:DMS$  or  $MS^-:SO_4^{2-}$  ratios different? Is the aerosol sufficiently acidic to drive off  $HNO_3$ ? (I strongly suspect that’s why you saw so little  $NO_3^-$ ; it was displaced to seasalt particles too big for the AMS to detect.)

The following paragraph was added, “The MSA to non-sea salt sulfate ratio ( $MSA:SO_4$ ) measured during ACE-ENA in the summer was 0.02 on average in the MBL (<1000 m), which is lower than historical estimates of the ratio. For example, Pszenny et al. (1990) reports the ratio as 0.05 in the North Atlantic in August-September, Berresheim et al. (1991) reports 0.033 in Western North Atlantic in September, Savoie et al. (2002) reports 0.06 in Bermuda in September

and 0.05 in Mace Head in August. Huebert et al. (1996) found 0.07 in marine air masses and 0.02 in continental air masses in June in the Azores. The measurements reported in those earlier studies are based on analysis of filter samples, and they may not be directly comparable to AMS measurements reported here. In particular, because the filter samples can measure larger particles than the AMS, this might suggest that some of the MSA was present on coarse aerosols, such as sea salt.”

I was also a bit surprised to see no estimates of how much of the aerosol in the boundary layer could have come from the free troposphere. Without level legs at the top of the MBL, you obviously cannot do a flux study, but you have tracers of long-distance transport that are immune to precipitation scavenging (CO, benzene, toluene) and at least 1 that is not (BC). Can you use those to put some bounds on how much of the  $\text{SO}_4^{2-}$  and organic aerosol came from above? Would that bring the  $\text{MS}^-:\text{SO}_4^{2-}$  in line with other work?

While this is a good suggestion for a follow-up study, accurate source apportionment is not straight-forward without chemical signatures that would correlate the aerosol with a source. Sulfate signatures in the AMS are generally the same for fresh and aged emissions, and while organics usually show some compositional diversity, a surprising observation in this campaign was uniformity of organic oxidation state (Figures 5 and 6). Adding a layer of complexity, it is expected that the structure of the MBL complicates simple application of back trajectories. Because of all these reasons, we found it appropriate not to speculate beyond the discussion in Figures 5 and 6.

The dramatic split in Fig. 6b in pHF is interesting. Since Table S1 says there were 5 profiles on 19 July, it seems likely that some profiles were in the putative fire plume while others were not. It might be worth trying multiple back trajectories on each of the profiles to establish a pattern like that seen in Clarke et al. (2013), where the ensemble of trajectories and their correlation with aerosol properties lent credibility to the trajectories over rather long distances.

Some of the ambiguity in the figure was removed by plotting only the vertical profile data in panels A and B. All of the vertical profiles happened to transect the plume. The figure in Clarke et al. (2013) looks very similar to Figure 14 in our paper. This is indeed what we were trying to do: generate multiple back-trajectories with systematic spatial offsets centered on one of the vertical profiles to make sure that we are not over-interpreting a single back-trajectory.

line 21 “fully” is meaningless. It suggests that all possibly relevant instrumentation was aboard, and that’s not possible even on much bigger aircraft.

We removed “fully” from this sentence.

line 26 average submicrometer non-refractory aerosol mass

We added “sub-micrometer” to this sentence

line 33 “1 % of the sulfate and no more than 3 % of the total aerosol” makes no sense. You presumably meant no more than 3 % of the submicron organic aerosol.

This sentence was revised to read, “MSA accounted for no more than 3% of the sub-micron, non-refractory aerosol in the boundary layer.”

line 140 Was there a typical altitude range of the spirals? “Through the atmosphere” is vague (and strictly speaking, incorrect). Was there a minimum altitude span criterion for inclusion in Table S1? Were there any criteria for where to do the spirals?

The spiral profiles were included in every flight to investigate the vertical structure. The altitude varied from flight to flight. Table S1 now includes the altitude information.

line 145 Was the standard aerodynamic lens used? What temperature was the vaporizer?

This information is now included, “The standard aerodynamic lens was used, and the AMS vaporizer was set to 600°C.”

line 155 Has the inlet efficiency for this inlet been characterized?

A characterization of the inlet has not been published. Unpublished data from the G-1 flight teams suggests the transmission efficiency is near 100% for particles smaller than 5 microns. The manufacturer’s manual on the inlet states that the inlet “transmit particles with diameters between 0.01 and 6 micrometers with better than 95% efficiency.” We added the following, “. The isokinetic inlet transmission efficiency is greater than 95% for 0.01 – 5 μm diameter particles.”

line 156 “switched ... based on cloud cover” seems unlikely. Surely switching was done based on whether the plane was actually in cloud.

This was revised, “Sample streams between two inlets were switched by the instrument operator aboard the aircraft based on the presence of clouds.”

lines 166–167 I’m a bit surprised at all the English (rather than SI) units. I guess that’s up to the journal editors. In addition, the OD of the tubing is irrelevant; it’s the ID that matters.

Stainless steel tubing is ¼” OD with .035” wall thickness. The Teflon is ¼” OD with 3/16” ID. The PEEK tubing is 1/16” OD with 0.055” ID. This was now re-stated, “The PTR-MS sampled air through a dedicated inlet that consisted of approximately 6" of 1/4" OD stainless steel with 0.035” wall thickness, followed by approximately 46" of 1/4" OD (3/16” ID) Teflon tubing, including a Teflon filter, and 36" of 1/16" OD (0.055” ID) PEEK tubing.”

line 186 The low supersaturation is presented inconsistently. Here and in Fig. S3 it is 0.1 %, while in Fig. 13 it is 0.13 %.

It should be 0.13% everywhere. This was now fixed.

line 173 I don't see how elevated DMS background in the summer IOP necessarily biases DMS measurements high. If it is indeed an isobaric interference (any idea what would do that?) then one would eliminate the overestimate by subtracting the background. If it was incomplete destruction of the DMS, then subtracting the background would produce an underestimate. If the interfering species was partly destroyed by the catalyst, then yes you would have an overestimate of DMS, but you could only claim that the patterns you see in DMS are accurate if the interfering species concentration was fairly constant.

We were unable to get consistent background measurements for DMS; they were consistently higher at the beginning and end of flights relative to in-flight. We attribute this to two factors 1) adsorption of an unknown interfering species on tubing walls at the airport followed by slow desorption of this interfering species during flight and 2) incomplete removal of the interfering species by the catalyst. These values were clearly observed to decrease as the aircraft transitioned to cleaner air. We choose low DMS values observed in the FT as the background. As a result, DMS values measured in the presence of the interference were biased high. On several flights, porpoising maneuvers into and out of the boundary layer showed clear and sharp transitions in the DMS measurements. On all flights, clear changes in DMS concentrations were observed when transitioning into the FT. Therefore, we feel the relative changes in DMS concentrations are instructive while the absolute concentrations are not. We note that changes were made to the inlet lines that greatly mitigated this interference in the winter campaign.

line 182 "more closely mimic" is incomplete. More closely than what?

"more" has been removed.

line 229–230 Has anyone ever claimed that MSA accounted for the majority of particulate sulfate mass?

This sentence was misleading and was removed.

line 235 The equation is only true if those species are the only acids and bases present in the aerosol. There are organic acids like oxalic acid and MSA. They are probably negligible here, but that ought to be noted.

We added, "The analysis also neglects organic acids, such as oxalic acid or MSA, but the concentrations of these in clean MBL are low. MSA, for example, was shown to account for only 1-2% of the non-refractory aerosol mass."

lines 242–245 Be more explicit about the use of the thermal denuder. Was there very little submicron seasalt (as is likely)? As determined by heating to what temperature? It's true that the AMS doesn't see coarse particles, so whether they are externally mixed seems irrelevant. Is there a claim here that there was little volatile material on coarse particles so the AMS wasn't actually missing significant  $MS^-$  and  $NO_3^-$ ?

The thermodenuder was placed in front of the FIMS instrument, which measured the particles size distribution from 10 to 500 nm, and heated to 300°C to remove non-refractory material from

the particles. These measurements indicated there was little submicron sea-salt and were meant to indicate as such. We have deleted the reference to the sea-salt being externally mixed. The sentence now reads, “Use of thermodenuder heated to 300°C in front of an aerosol sizing instrument aboard the G-1 aircraft during the ACE-ENA campaign was used to infer that the AMS observations are of non-sea salt submicron marine aerosol.”

line 250 It’s not wrong, but a bit odd to use a moving average with an even number of points, meaning that the time represented by the average is between the times of two data points, but not right on them. I suppose it doesn’t matter here since there aren’t any comparisons here that depend on close synchronization.

It appears that Igor Pro defaults to an odd number of points when an even number of points is specified in the smoothing function. We changed the number of points in the text to 11.

line 275 Acid-catalyzed reactive uptake of organic vapors is an interesting idea. Any citations for it? Could SOA production via that mechanism be fast enough to account for the extra organic aerosol in the MBL?

Acid-catalyzed uptake of IEPOX has been shown to be more efficient at SOA production than non-IEPOX photochemical reactions. Some of the relevant literature includes Surratt et al. (2010), Lin et al. (2012) and D’Ambro et al. (2019). We added the following, “While the isoprene concentrations measured during ACE-ENA were low and close to the detection limit of 0.1 ppb (Table 2) SOA formation from acid-catalyzed IEPOX chemistry has been shown to be significantly more efficient than from non-IEPOX photochemical mechanisms (Surratt et al., 2010).”

line 371 (This is a hobbyhorse of mine.) Airmasses do not have origins! The air always came from somewhere earlier and has traces of that left in it. If there was near-total scavenging event or a large influx of pollution that dwarfs whatever was present, then one could claim there is an origin of the characteristics of the air mass. Ascribing an origin to an air parcel in the MBL is particularly absurd, since there is almost always entrainment mixing going on meaning that much of the air was in the FT within the last few days. Simple back trajectories are not really capable of conveying that.

This was rephrased, “Supplementary Figure S7 corroborates the low-altitude air masses as strongly influenced by marine conditions.”

line 402 Surely you mean ”summer and winter” rather than ”summer and fall”.

Yes, this has been fixed.

Fig. 4 It’s jarring that the pie charts for the organic fraction of the aerosol are larger than the the total aerosol.

We have resized the organic pie charts.

Fig. 6 This is an interesting figure, but the caption isn't as clear as it ought to be. It appears that in panels A and B, the organic loading is averaged into 100 m bins, while the pH<sub>f</sub> is for individual (10 s?) averages. The pH<sub>f</sub> data in panel B has a remarkable split between nearly neutralized and very acidic aerosol, as though some of the 5 spirals that day were in the pollution plume while the others were not. Of course the averaged organic loading cannot show that. Would it be worth plotting the spirals separately, or grouped as plume vs. non-plume? Were panels C, D, and E for the entire vertical profile? What does "normalized" mean here? Same area under the curve? It didn't happen often, but it appears from panel C that MS<sup>-</sup> was sometimes 10 % to 15 % of the SO<sub>4</sub><sup>2-</sup>. Is that real?

The reviewer is right that there was ambiguity in panels A and B of this figure. The organic loadings were plotted only from vertical profiles, but the pH<sub>f</sub> values were plotted from the whole flight, including the level legs. This was fixed so that both the loadings and pH<sub>f</sub> values are from the vertical profiles. All the spirals sampled the plume, but the level legs did not.

Fig. 14 While the maps are quite pretty, there is a lot of information, such as bathymetry, that is unimportant to the paper. I don't actually object much to that even though it is best practice to exclude irrelevant material from graphics. However, including the political divisions within countries is clearly excessive.

We removed the political boundaries from the figure.

**Equations in the supplement** I assume the Copernicus editors will help you figure out what should be italicized and what should not be.

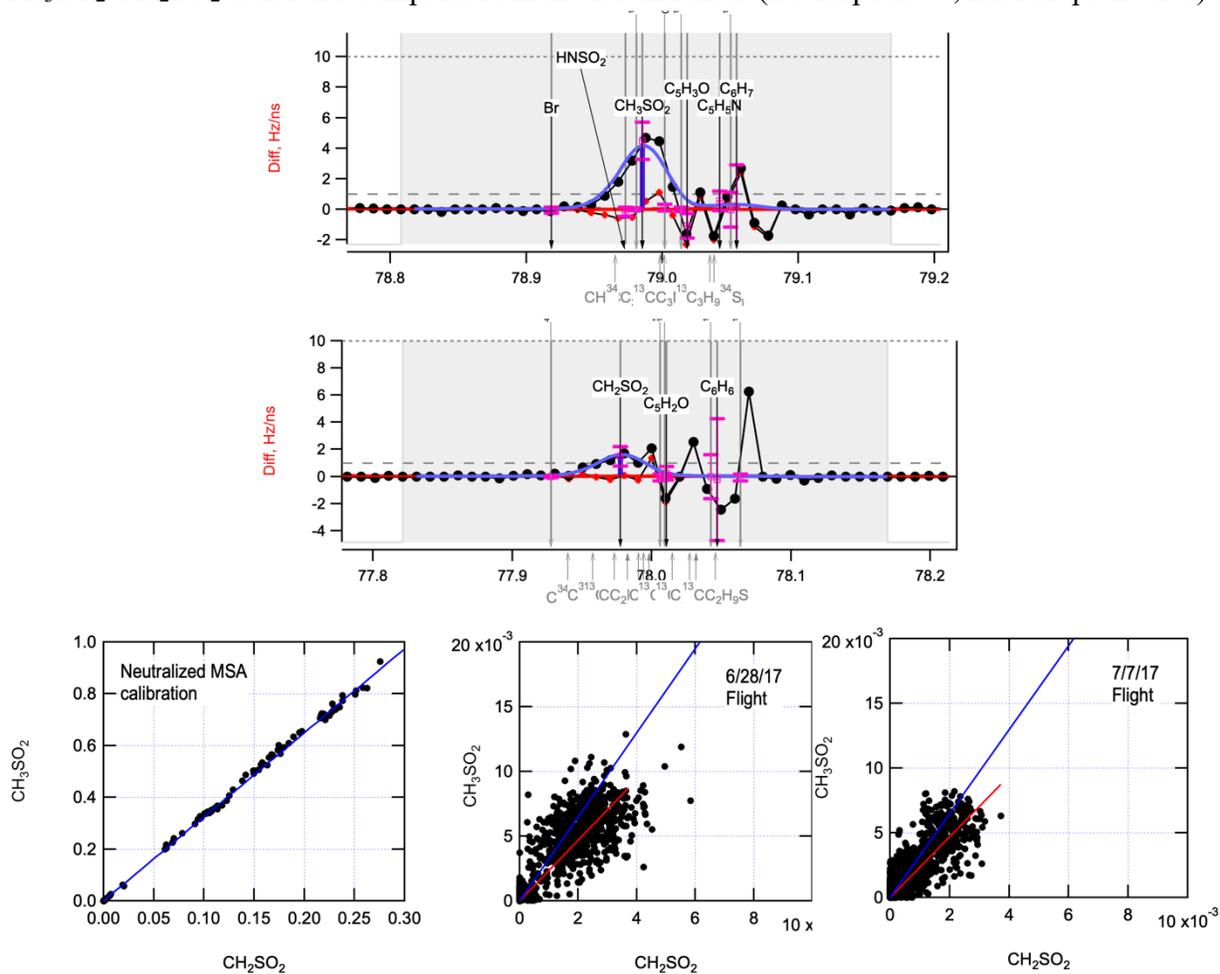
We will follow the editor's guidance on formatting.

**CE of MSA in the supplement** This is a misinterpretation of Middlebrook et al. (2012). It's not the pH that matters—it's whether the aerosol is liquid or solid. That said, since MSA salts are solid and MSA itself is liquid (much like H<sub>2</sub>SO<sub>4</sub> and salts thereof), the CEs you propose are reasonable in the lab. I'm not sure what you're doing with the field data—the particles are presumably internal mixtures with only small contributions from MS<sup>-</sup>. In that case, it's the presence of liquid H<sub>2</sub>SO<sub>4</sub> that will determine CE for the entire aerosol.

This was re-stated as follows, "This is the largest source of uncertainty in translating the calibrations to ambient measurements of MSA, as ambient MSA is more acidic than the neutralized laboratory MSA, which implies that the ambient particles less viscous, which is expected to affect CE (Middlebrook et al., 2012)." It is indeed true that the presence of sulfuric acid determines the CE of ambient measurements and that's why we are using CE = 1 for all ambient data.

**PIKA vs Squirrel** Did you get ICH<sub>3</sub>SO<sub>2</sub> + , ICH<sub>2</sub>SO<sub>2</sub> + and ICH<sub>4</sub>SO<sub>3</sub> + from PIKA or did you use unit mass data from Squirrel? I guess you did it with PIKA, which would make sense, particularly in the field, where other species would be present at those unit masses. To look for interferences, it might be useful to plot ICH<sub>4</sub>SO<sub>3</sub> + vs ICH<sub>3</sub>SO<sub>2</sub> + and ICH<sub>2</sub>SO<sub>2</sub> + vs ICH<sub>4</sub>SO<sub>3</sub> + to see whether you have the same fragmentation pattern in the field as you had in the lab.

We quantified these ions by fitting them in PIKA, as shown below. Evaluation of some of the individual HR fits suggests that there are no major interfering ions, especially in very clean conditions. In the field, these ions are of very low intensity, corresponding to low MSA concentrations, which makes comparisons to lab fragmentation patterns challenging. The plots below for June 28 and July 7 flight, when MSA was especially abundant suggest that the  $\text{CH}_3\text{SO}_2/\text{CH}_2\text{SO}_2$  ratios are comparable in the lab and field (lab slope is  $\sim 3$ , field slope is  $\sim 2.5$ ).



**Table S1** Including 4 digits after the decimal point for latitude and longitude specifies the point to within 11 m. Seems excessive. Might be useful to add an altitude range, unless that was constant (in which case I'd like to see that somewhere).

The latitude and longitude information was rounded up and altitude is now included.

**Fig. S3** The y axes on panels E and F are labeled  $\text{SO}_4$  rather than  $\text{NH}_4$ .

The error was fixed.

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