Interactive comment on "High Summertime Aerosol Organic Functional Group Concentrations from Marine and Seabird Sources at Ross Island, Antarctica, during AWARE" by Jun Liu et al.

Anonymous Referee #2

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The current paper reports organic concentration and speciation of aerosol organic functional groups during AWARE. There is a lack of measurements in the study region and these measurements are very timely. However, the paper is poorly written and it is not recommended for publication in ACP. A more specialized lower impact factor journal is recommended.

We thank the referee for reviewing the manuscript and recognizing the importance of our unique, first-of-their-kind, yearlong measurements of organic aerosol mass and functional group composition. **(Page and line numbers in this response reference the location in the discussion paper where the text is inserted. The revised manuscript will note these revisions separately with tracked changes when it is posted.)**

Our findings are appropriate for ACP rather than a lower-impact journal because they meet the journal requirements, which are: "Research articles report substantial new results and conclusions from scientific investigations of atmospheric properties, and processes within the scope of the journal. Please note that the journal scope is focused on studies with general implications for atmospheric science rather than investigations which are primarily of local interest."(https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html)
Specifically the atmospheric properties investigated in this manuscript were organic aerosol composition and mass emitted from marine and sea bird sources in Antarctica. Three main results are

1) OM and PM concentration were 5-150 times higher during summer than winter, with summer OM mostly from sea bird sources.
2) Summertime OM has amide group characteristic of seabird emissions.
3) Carboxylic acid group from natural sources were from secondary formation pathways.

These results are new because there are no published reports of any of these results. While Result (1) is consistent with seasonal trends for CN (Hogan and Barnard, 1978; Parungo et al., 1981; Bodhaine, 1983; Gras, 1993; Kim et al., 2017) and PM (Wagenbach et al., 1998; Jourdain and Legrand, 2002; Weller and Wagenbach, 2007; Udisti et al., 2012; Legrand et al., 2017a; Legrand et al., 2017b; Asmi et al., 2018) , prior results did not include OM. Similarly Result (2) is consistent with reports attributing some CHN and CHNO fragments to seabirds (Schmale et al., 2013) , but our results are the first to provide their contribution to PM and summertime maximum. Result (3) is consistent with observations of marine SOA (Frossard et al., 2014) but new in that this is the first study showing a coastal (i.e. continental not marine) source of SOA in Antarctica.
These results are substantial because there are no other measurements of seasonal OM concentration in all of Antarctica, which makes calculation of radiative fluxes in this entire region very uncertain. In addition there is no prior evidence of an OM functional group marker for seabird sources, which has prevented the identification of the

contribution of seabird sources worldwide. Finally, the lack of characterization of secondary particle sources in Antarctica means that this important pathway has not been included in models of aerosol formation and growth over Antarctica.

The reasons why this study is within the scope of the journal are:

1) The following past ACP publications of Antarctic aerosol measurements show that this topic is well within the scope of the journal: Giordano et al., 2017; Asmi et al., 2010; O'Shea et al., 2017; Legrand et al., 2017a; Legrand et al., 2017b; Hara et al., 2014; Hara et al., 2013.
2) Two specific ACP publications with results similar to ours are described below:
    a. Legrand et al. (2017a, b) reported multiple year size-segregated aerosol composition in central Antarctica (Concordia) and found chloride depletion as well as different seasonal patterns of $nssSO_4$ and MSA characterizing to particles in central Antarctica. Our year-round measurements at McMurdo Station has a similar scope to these two publications except that we measured organic functional groups in addition to inorganics and that our main results are about OM rather than sulfate.
    b. Giordano et al. (2017) reported aerosol measurements by AMS near McMurdo Station during October–December 2014 and August-October 2015 with source apportionment by PMF. The study found that sulfate aerosol mode was a large fraction of non-refractory submicron particle mass, even though the mass concentrations were not corrected for collection efficiency in two non-consecutive summer and spring months. Our year-round measurements at McMurdo Station have a similar scope to this publication except that we measured by FTIR and XRF rather than AMS and that our main results are about OM rather than sulfate.

3) Our study has general implications rather than just local interest for the following reasons:
    a. Because there are so few other measurements on the entire Antarctic continent, the closest other station with aerosol measurements is 300 km away. In fact, McMurdo Station is one of the two sites that have published aerosol measurements starting in 1968, with the other one being the Amundsen Scott Station at the South Pole. The site has at least 10 publications describing aerosol measurements over the past 50 years, most of which were limited to summer (Cadle et al., 1968; Warburton, 1973; Ondov et al., 1973; Hogan, 1975; Hofmann, 1988; Hansen et al., 2001; Mazzera et al., 2001a; Mazzera et al., 2001b; Giordano et al., 2017; Kalnajs et al., 2013; Khan et al., 2018) . Moreover McMurdo is the only station in the Ross Ice Shelf region.
    b. Marine and seabird sources are common along the coastlines of the continent of Antarctica, so the results from this study apply to much of coastal Antarctica which covers hundreds of square miles.

c. Specifically for the Crozier colony, we estimate the plume from seabirds to increase OM by 20 times for an area that covers 1000 km downwind with an average width of 50 km. This calculation is consistent with the 4 month summertime seabird-related OM concentrations of 0.16 μg m^{-3} compared to winter of 0.001 μg m^{-3} at McMurdo, which is >70 km downwind.
d. Since this increased OM causes submicron PM concentrations to increase 20% because of seabird emissions for such a large area, the direct and indirect effects of aerosols will be increased in these regions. For example, in the Arctic, the indirect effect of seabirds was shown to change by -0.5 W m^{-2} pan-Arctic radiative forcing averaged over the 14,000,000 km^2 of the Arctic Ocean (Croft et al. (2016) .

We recognize the referee's concern that the manuscript is poorly written but we note that Referee #3 describes the work as well written. Nonetheless we recognize that there are always ways to improve written communication that is intended for diverse audiences so, in accordance with the more specific suggestions of Referee #1, we have improved the manuscript by the reorganization of the introduction, expansion of the discussion of relevant literature, improvements in transitions between topics, clarification of the language, and minor grammatical corrections, as described below:

1. Reorganization of the introduction:
a) Moved season definition and seasonal meteorological conditions from P4 Line 25 to section 2 P3 Line 11.
b) Moved description of PMF and K-means clustering operation to Method P3 Line 25 P5 Line15 (as described in response to Referee #1).
c) Improved organization of the fourth paragraph in section 3 P5 Line 23-33 (as described in response to Referee #1).

2. Improvements in transitions between topics (as described in response to Referee #1):
a) Rephrased P1 Line 30-31.
b) Rephrased P3 Line 9.
c) Rephrased P5 Line 1-22.

3. Expansion of the discussion of relevant literature (as described in response to Referee #1):
a) Added discussion of source of sea salt aerosols at P6 Line 6.
b) Added comparison to Arctic OM at P6 Line10.
c) Added summary of reasons for attribution of FTIR PMF factor to seabird and marine sources in at P8 Line 6.

4. Clarification of the language and methods.
a) Added more detailed about the measurement setup. (P3 Line 20) (as described in response to Referee #1).
b) Clarified OM calculation and detection limit for FTIR measurement (P4 Line 3 and P7 Line 19). (as described in response to Referee #1).

c) Clarified why the CN concentrations that remain after SLCE (spikes) are removed are considered representative of the natural background rather than local pollution from McMurdo (P4 Line 25) (as described in response to Referee #1).
d) Clarified the marine and FFC contributions to amine (P7 Line 22). (as described in response to Referee #1).
e) Changed units from imperial to metric "... less than 100km downwind..." (P7 Line 30)

5. Minor and grammatical corrections:
a) Rephrased "... modeling estimated that.." to "... modeling was used to estimate that... " (P2 Line 9).
b) Rephrased "Biological emissions from marine sulfate sources" to "Sulfate from marine biological... " (P2 Line 20).
c) Rephrased "The organic composition of particles in marine and Arctic regions..." to "For comparison, in marine and Arctic regions, the organic composition of particles..." (P2 Line 22).
d) Corrected abbreviation from "CNH and CNHO" to "CHN and CHNO" (P2 Line 32).
e) Added description of location of Cape Crozier (P7 Line 33).
f) Rephrased "more regionally-representative patterns " to "..regionally-representative or “background” concentrations " (P8 Line 21).
g) Rephrased sentence "...the FFC factor had a higher concentration than M&S in winter but the concentrations were so low that the quantification of the M&S factor in winter is very uncertain." (P8 Line 31).
h) Fixed capitalization"... M&S factor..." (P9 Line 2).
i) Rephrased "seabird" to "seabird-related" (P9 Line 3).

Major issues:
- The paper does not present any introduction of the aerosol composition in Antarctica. Mostly, the introduction seems a report of the history of a specific station.

The Introduction of the original posted ACPD paper summarizes the measured aerosol composition and concentration at McMurdo Station as well as the few existing hygroscopicity and organic aerosol measurements in all of Antarctica. We agree that the Introduction could be expanded to provide

1) a description of the paucity of nearby stations and the lack of long-term (more than one year) organics measurements (see item a) below).
2) a more thorough review of the limited study of seasonal trends in Antarctica (see items b) and c) below).
3) a more thorough review of the detailed OM measurements in Antarctica (see items d) and e) below).

These changes are itemized below:
a) Start of the second paragraph:
"Since McMurdo Station is the only site with measurements of PM, EC, OC, number concentrations that is within 300 km of the Ross Ice Shelf (which covers an area of more than 500,000 km^2). Furthermore, the station is unique in that McMurdo Station is one of the two sites that have published aerosol measurements starting in 1968, with the other

one being the Amundsen Scott Station at the South Pole. The site has at least 10 publications describing aerosol measurements over the past 50 years, most of which were limited to summer (Cadle et al., 1968; Warburton, 1973; Ondov et al., 1973; Hogan, 1975; Hofmann, 1988; Hansen et al., 2001; Mazzera et al., 2001a; Mazzera et al., 2001b; Giordano et al., 2017; Kalnajs et al., 2013; Khan et al., 2018) . No stations in Antarctica measured inorganic chemical composition year-round until 1978 (Parungo et al., 1981) , and none have measured year-round organic components. " (P2 Line3)

b) End of the second paragraph:
"Many measurement campaigns were limited to austral summer months because of restrictions on access (Cadle et al., 1968; Ondov et al., 1973; Warburton, 1973) and so lack information on seasonal changes." (P2 Line21)

c) After the second paragraph:
"The few year-round aerosol concentration and composition measurements in Antarctica were collected at several sites in coastal Antarctica (all of which are more than 1500 km from McMurdo Station) (Hara et al., 2005; Wagenbach et al., 1998; Jourdain and Legrand, 2002; Gras, 1993; Hara et al., 2004; Hara et al., 2010; Weller et al., 2013; Minikin et al., 1998; Read et al., 2008) and at several sites on the Antarctic Peninsula (more than 3000 km from McMurdo Station) (Asmi et al., 2018; Mishra et al., 2004; Kim et al., 2017; Saxena and Ruggiero, 1990; Savoie et al., 1993; Loureiro et al., 1992) , as well as at the South Pole (more than 1000 km from McMurdo Station) (Hansen et al., 1988; Bodhaine et al., 1986; Harder et al., 2000; Parungo et al., 1981; Bodhaine, 1983; Hogan and Barnard, 1978) and at Dome C (more than 1000 km from McMurdo Station) (Legrand et al., 2017b; Legrand et al., 2017a; Udisti et al., 2012) . At the South Pole, aerosol particle number concentration ranged from 10 to 30 cm^{-3} in winter and 100 to 300 cm^{-3} in summer (Bodhaine, 1983; Parungo et al., 1981; Hogan and Barnard, 1978) . This low winter and high summer seasonal difference has been observed also at coastal Antarctic sites, but the average concentrations were typically higher with summertime concentrations ranging from 300 to 2000 cm^{-3} and wintertime concentrations from 10 to 200 cm^{-3} (Kim et al., 2017; Gras, 1993) . Consistent with this seasonal difference in particle number concentrations, most summertime non-sea salt sulfate mass concentrations were at least 5 times higher than winter concentrations (Jourdain and Legrand, 2002; Weller and Wagenbach, 2007; Udisti et al., 2012; Legrand et al., 2017a; Asmi et al., 2018) , likely because of the contributions from biogenic DMS emissions from the surrounding Southern Ocean. However, most sea salt aerosols had wintertime maximum concentrations with more than two times more Na^+ mass concentrations in winter than summer (Parungo et al., 1981; Wagenbach et al., 1998; Jourdain and Legrand, 2002; Weller and Wagenbach, 2007; Jourdain et al., 2008; Udisti et al., 2012; Legrand et al., 2017b; Legrand et al., 2017a; Asmi et al., 2018) " (P3 Line1)

d) After the old P2 Line 22 line:
Three sentences about organic nitrogen were revised as suggested by Referee #1. (as described in response to Referee #1).

e) Before the old P2 line 29:

“Sugar, levoglucosan, phenols and anthropogenic persistent organic compounds were measured in ambient aerosols at Mario Zucchelli Station and Concordia Station (Zangrando et al., 2016; Barbaro et al., 2016; Barbaro et al., 2017; Barbaro et al., 2015) Carboxylic acids with low molecular weights were also measured at Mario Zucchelli Station, Concordia Station, and Dumont d'Urville (Barbaro et al., 2017; Legrand et al., 2012)

- Scientific questions and objectives are really not presented, neither discussed or summarized.

The major scientific questions and the corresponding three main results are presented, discussed, and summarized in the discussion paper at the following lines:
1) The question of how the concentration and composition of OM varies seasonally is presented on P1 at Line 20 in the abstract and on P3 at Line 3-4 in the introduction; it is discussed on P7 at Lines 10-21 in section 4; and it is summarized on P8 at Line27-28 in the conclusions.

2) The question of what the contributions from natural sources of OM are is presented on P1 at Line 21 and on P1 at Line 25 in the abstract and on P3 at Line 5-7 in the introduction; it is discussed on P7 at Line 10-35 and on P8 at Line 1-6 in section 4; and it is summarized on P8 at Line 29-31 and on P9 at Line 1-3 in the conclusions.

3) The question of whether or not there is a secondary pathway that contributes to OM formation is presented on P1 at Line 25-26 in the abstract and on P3 at Line 8-9 in the introduction; it is discussed on P8 at Line 7-17 in section 4; and it is summarized on P9 at Line3-5 in the conclusions.

In addition, we have now revised part of the abstract and introduction so that these scientific questions can be more easily identified by readers as follows:

In abstract: "Observations of the organic components of the natural aerosol are scarce in Antarctica, which prevent us from better understanding natural aerosols and their connection to seasonal and spatial patterns of cloud albedo in the region."

In the introduction: "This manuscript characterizes the sources of organic aerosol across four seasons in Antarctica. Dust, sea salt, and non-sea salt sulfate mass concentrations measured by XRF are used to separate the seasonal contributions to inorganic particle components. Seasonal patterns of natural marine and coastal-sourced organic aerosol are identified from the functional groups after separation of local emissions. " (P3 Line9)

- After reading a paper a number of times, and looking at the figures, one can argue the main results are the impact of sea birds and marine sources in Antarctica. Not surprisingly at all, the reader does not understand if this is simply a bad measurement site (bad luck) or if the study has any implication.

We agree with the Reviewer that one of the three main results of this paper is the contribution of seabird emissions to summertime submicron particles. We further note that this result is important because of the reasons noted above for why McMurdo Station is not only a good and representative sampling site but also the home of one of the earliest aerosol measurements on the continent.

We have added the following text to the discussion to highlight the potential regional implications of seabird emissions based on results for the Arctic:
"The emissions from seabirds have significant regional implications in polar areas due to because of their large population and wide distribution (Croft et al., 2016; Riddick et al., 2012) Chemical transport model simulations suggest that emissions of reduced nitrogen from seabirds in the Arctic could significantly increase aerosol particle formation, and in turn cloud droplet number concentration and cloud albedo, yielding as much as -0.5 W m^{-2} radiative forcing averaged over the 14,000,000 km^2 of the Arctic Ocean (Croft et al., 2016) ."

In addition, to make a simple approximation of the impact of seabird plumes in Antarctica (see Appendix), we use a simple Gaussian dispersion model to estimate that the 80,000 Cape Crozier seabird breeding pairs will affect an area of approximately 50,000 km^2 by increasing aerosol optical by 40% (from 0.010 to 0.014). Scaling this up to all Adelie penguins in Antarctica (2 to 3 million breeding pairs) and assuming not overlapping plumes of similar dispersion, we estimate that as much as 4,000,000 km^2 could be affected by the approximately 200 seabird colonies along the Antarctic coastline (Borowicz et al., 2018; Ainley, 2002; Knox, 2006) .

I am afraid I cannot be any positive at this stage.

We are very sorry that the referee “cannot be any positive” at this stage. The coauthors on this manuscript request that the Referee reconsider his/her opinion based on the revisions presented here. Specifically we argue that the manuscript is appropriate for ACP since we are presenting substantial and new results with general implications. We thank the referee for reconsideration and we are happy to provide additional responses and clarification if needed.

Appendix

1. Plume Dispersion:

The following equation is used to calculate the concentration (C) from dispersion of the plume from the seabird colony emissions:

$$C(x,y,z;H) = \frac{Q}{\sqrt{2\pi}LU\sigma_y} exp[-\frac{1}{2}(\frac{y}{\sigma_y})^2]$$ (Turner, 1994)

Table A1. Parameters for plume calculation.

Parameters	Description	Value
x	horizontal distance along the wind direction	calculated
y	horizontal distance normal to the wind direction	calculated
z	vertical distance from the source	calculated
L	boundary layer height	400 m
H	height of emission source	0 m
Q	emission strength of the point source	6×10^6 μg s^{-1}
σ_y*	dispersion coefficient in y direction, determined from measured atmospheric conditions	814 m

*σ_y was determined for stability class C for the average wind speed of 6 m s^{-1} and radiation measured at McMurdo Station (Martin, 1976) . (Pasquill, 1961)

This is a simplified Gaussian plume dispersion model for capped inversion. We assumed that emission was from a point source with zero elevation at Crozier and ignored particle sinks during transport. Boundary layer height was calculated by the ARM standard method (Liu and Liang, 2010) from sounding profiles measured during AWARE, giving a summertime average boundary layer height of 400 m (Table A1). We used a constant wind speed of 6 m s^{-1} and direction from Cape Crozier to McMurdo Station. Fitting this equatio to the observed average summer concentration of 0.16 μg m^{-3} OM at McMurdo Station gives Q of 6×10^6 μg s^{-1} for OM. To estimate the ammonium contribution to PM, the 80,000 breeding pairs at Crozier (Lyver et al., 2014) have an ammonia emission strength of 2×10^6 μg s^{-1} from equation (2) (Riddick et al., 2012) with the estimated coefficients for the nitrogen metabolism in that study. If all of the ammonia is taken up by submicron particles then the ammonium concentration from seabirds at McMurdo Station is estimated to be 0.05 μg m^{-3}, which is reasonable given the measured submicron sulfate mass concentration. Using the Q calculated for OM and defining the plume to be the area for which OM exceeds 0.02 μg m^{-3} (20 times higher than the wintertime natural OM average of 0.001 μg m^{-3}), we find the plume has an estimated length of 1000 km with an average width of 50 km, giving an area of 50,000 km^2 with average OM concentration of 0.04 μg m^{-3}. Including the ammonium concentration for the same plume area adds an additional 0.01μg m^{-3} to PM1, making a total of 0.05 μg m^{-3} from seabirds for the Crozier plume.

2. Extinction:

The background AOD at 500 nm (Cimel Sunphotometer) was determined to be 0.05 by using the average of minimum values from 18 November 2015 to 29 February 2016 (https://www.arm.gov/research/campaigns/amf2015aware). This number is consistent with measurements of median aerosol optical depth (500 nm) of total aerosol, which ranged from 0.02 to 0.06 at 9 stations across Antarctica (Tomasi et al., 2007) . To estimate the fraction of AOD that is from submicron scattering, we scale the AOD by the ratio of PM_1 to PM_{10} scattering (2.3/3.3) to get 0.035 (Table A2). Of this, we can estimate the fraction associated with seabirds by scaling by the ratio of seabird-related PM_1 to PM_1 (0.21/0.52) to get 0.014 at McMurdo Station. The seabird-related part of

AOD for the entire plume spread over 50000 km^2 is then one-fourth of this (0.05/0.20), at 0.004.

If the 80,000 seabird breeding pairs at Cape Crozier affect an area of approximately 50,000 km^2 by increasing aerosol optical by 40% (from 0.010 to 0.014), then the 2 to 3 million penguin breeding pairs in Antarctica (assuming not overlapping plumes of similar dispersion) will affect as much as 4,000,000 km^2 (Borowicz et al., 2018; Ainley, 2002; Knox, 2006) (Table A3).

Table A2. Estimate of seabird-related AOD from the Crozier plume.

Quantity	McMurdo Station (70 km downwind)	Crozier Plume (Averaged over 50000 km^2)
PM_1 scattering coefficient	2.3 1/Mm	
PM_{10} scattering coefficient	3.3 1/Mm	
PM_1 (seabird-related)	0.21 $\mu g\ m^{-3}$	0.05 $\mu g\ m^{-3}$
PM_1*	0.52 $\mu g\ m^{-3}$	0.20 $\mu g\ m^{-3}$
AOD total	0.050	-
AOD submicron	0.035	
AOD submicron seabird	0.014	0.004

*PM_1 is calculated as the sum of the measured OM, NSS sulfate, and sea salt, plus the ammonium mass concentration from the Crozier plume calculated from the Riddick et al. (2012) source and diluted to the distance of McMurdo Station (70 km).

Table A3. Estimate of area influenced by Adelie penguin emission.

	Penguin Breeding Pairs	Potential Plume Area
Cape Crozier	80,000	50,000 km^2
Antarctica	2 million to 3 million	1 to 4 million km^2

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