Response to Reviewer comments

We thank the reviewer for their supportive and constructive comments on the manuscript. We feel that the paper has been improved by the review process. Below, we address each of the reviewer's specific and technical line-by-line comments. The reviewer comments are in black text while the responses are in *blue italics.*

General Comments

In this paper, the authors present stable nitrogen and oxygen isotope measurements of aerosol nitrate collected on filter samples during 2018 and 2019 in the Southern Ocean between South Africa and Antarctica, and in the Weddell Sea. The measurements are then analyzed based on previous literature-based fractionation and isotope information on both nitrate nitrogen and oxygen sources in the different key study regions, including anthropogenic NOx, lightning NOx, emissions of RONO2, and snow-NOx emission, as well as O3 and other oxygen sources and equilibrium impacts. The introduction is thorough and well-written, and presents a clear context for the findings described in following sections. I find this paper and its findings compelling, and certainly worthy of publication, but there are a number of issues that need to be addressed prior to publication.

As well, there are a fair number of typographical errors and style guide recommendations that need to be addressed, and I would encourage the authors to take greater care in the future to address these details prior to manuscript submission, in particular for journals where there is no typesetting prior to the review process.

Specific Comments

Line 95 – Technically, É□ = (KIE − 1) × 1000‰, where KIE, the kinetic isotope effect is the ratio of the rates, = k_(primary N isotopologue)/k_(stable 15N isotopologue), so É› is the ratio – 1 × 1000‰.

This technicality will be amended in the text. The sentence on line 95 will read "In contrast, snow emitted NO_x typically has a very low $\delta^{15}N$ signature due to the large fractionation ($^{15}\varepsilon$) of ~ - 48‰ (Berhanu et al., 2014 and 2015) associated with NO_3^- photolysis in the snowpack, where $^{15}\varepsilon$ = (KIE - 1) x 1000‰ and the kinetic isotope effect (KIE) is the ratio of the rates with which the two isotopes of N are converted from reactant to product."

Line 142 – the date range could be simplified by writing 7-19 December 2018. Similarly, on Line 143, you could write 4 January to 21 February 2019. Also, at some point, and this is probably the ideal time, it should be stated what time zone (UTC? local time? South Africa Standard Time?) is being used to describe the dates for the legs and the times for the filter sample collections.

Both date ranges will be edited in the text. The time zone used to describe the dates for the cruise legs and the times for the filter sample collections was GMT. This will be included in the text as recommended by the reviewer.

Lines 161-162 – Were the samples that were taken for less than 24 hours due to stagnant conditions due to unusual ship manoeuvres? Or was it a combination of both stagnant conditions and unusual ship manoevres? Also, Table S1 lists "Daylight (hrs)" (which should be "hours" or "h") but not total sampling duration times. So the reader isn't left doing their own calculations based on the location and time of year relative to the equinox, perhaps the authors could include both "total daylight hours sampled" and "total sampling duration" in Table S1. It would also be good to include sampling start and end times (with a reference time zone) in the table, rather than just the start day and stop day, to demonstrate consistency with the 13-88 hours reported in Line 162.

Samples that were taken for less than 24 hours were done so if either the ship was stationary, resulting in stagnant conditions, or if unusual ship manoeuvres occurred, as both could have potentially led to contamination of our samples. This will be clarified in the text.

Table S1 will be amended to include the sampling duration in hours (h) to demonstrate consistency with the 13 to 88 hours reported in Line 162. To clarify, the daylight hours are the number of daylight hours that were available during the time of sampling, not the number of daylight hours that we explicitly sampled. We do not refer to this data in the main text and have therefore decided to remove this column for simplicity and to avoid confusion.

The high-volume air sampler was controlled by a sector collector, therefore the pump would only switch on when the wind was blowing from the front of the ship for a certain amount of time at a certain speed, as outlined in the methods. This will be reiterated in section 4.1 to remind the reader. As a result, the high-volume air sampler was not operating for the entire duration of every filter deployment and the sampling duration is not equivalent to subtracting the end times from the start times. Instead, the sampling duration was determined by the difference in the pump's hour meter reading before and after a filter sample deployment. A footnote will be added to supplementary Table 1 to clarify this.
Considering this and the fact the many of the filter deployments span multiple days, we do not think that it is necessary to include the time at which each sample was deployed and collected.

Line 168 – for consistency with line 164 "field blank", you could write "field blank filters" or "field blank set of filters".

This will be edited to read "field blank filters" as recommended.

Lines 175-183 – I have questions about the filters. What is the precision of the IC system analysis? Is it 0.3 µmol/L? What was the reasoning behind the other analysis, and why was this only done on a subset of the filters? In line 183, the authors suggest the average [NO3-] is "reported", but where? Maybe point to it here? And why not report both? And are they labeled as this or that or both? There should be a notation in Table S3 for the samples that were analyzed using both methods.

The precision of the IC system analysis is 0.3 µmol L^{-1}. This will be clarified in the text.

Our initial plan was to measure all of our samples using ion chromatography (IC) to determine nitrate concentration as well as other inorganic ion concentrations that are outside the scope of this manuscript. However, at the time of our sample analyses the IC was not operational. To prevent any

delays in analysis of the samples for isotopic composition, we decided to use the Lachat QuikChem® flow injection autoanalyzer to determine the nitrate concentrations. This technique does not however provide information on any of the other potentially important inorganic ions (i.e., ammonium, sulfate, sodium, chloride, etc.), which are not reported here but will be used in other studies.

Once the IC system was operational, we performed a second separate series of filter extractions using the left-over filter samples to measure the other cations and anions inclusive of nitrate. We thus thought it most accurate to report an average of the two measurement techniques. We did not report both nitrate concentrations measured using each technique separately, as we did not see a methods comparison to be pertinent to this manuscript, especially considering that the average value was used in all further data analysis.

Furthermore, separate filter extractions were used for each measurement technique, such that any difference in concentration is likely driven by heterogeneity on the filter as opposed to the technique used to measure the extract concentration.

Our methods inaccurately state that a subset of aerosol samples was analysed for nitrate concentration using the Lachat QuikChem® flow injection autoanalyzer, when in fact all samples were measured using both methods. The methods section will be amended to reflect this correctly. We will also refer to the table when the nitrate concentration is reported in the text.

Furthermore, a conversion error was found in the determination of the volume of air sampled during each filter deployment. This only effects the atmospheric NO_3^- concentration (ng m^{-3}) and not the isotope data. The atmospheric NO_3^- concentration is reduced compared to the original manuscript; however, the same trends are observed with latitude. We have updated the necessary figures, tables, and online data repository.

Line 198 – Where are the seawater samples provided? If you're not providing a table or documentation on the seawater sample data, it would be prudent to provide a link at this point, and not just at the end of the manuscript. Also, what depth does "position at depth ± 5 m" imply? Was the depth location actually uncertain to within a 10 m range? Lastly, the reported data repository on line 500 is an incomplete link that goes nowhere, so this needs to be sorted out prior to publication, or a final review of the paper.

The data link has been updated: https://doi.org/10.5281/zenodo.5006983
We will edit the text to reflect the depth of ships underway system as "approximately 5 m", because the depth at which the underway inlet sits is not actually uncertain to within a 10 m range as queried by the reviewer.

We will also include a supplementary table that highlights the seawater data as shown below.

Table SX: The average (Avg) and standard deviation (SD) sea surface nitrite concentration ($[NO_2^-]$ (µmol L^{-1})) measured during the early summer (ES) and late summer (LS) cruise transects. The date (dd/mm/yyyy), time (GMT), latitude (° S) and longitude (° E) of each sample is also given.

Leg	*Date*	*Time*	*Latitude*	*Longitude*	*$[NO_2^-]$ (µmol L^{-1})*	
	(dd/mm/yyyy)	*(GMT)*	*(° S)*	*(° E)*	*Avg*	*SD*
ES	*07/12/2018*	*10:00:00*	*34.23*	*17.85*	*0.13*	*0.03*

ES	*07/12/2018*	*14:00:00*	*34.50*	*17.09*	*0.11*	*0.01*
ES	*07/12/2018*	*18:00:00*	*34.50*	*16.17*	*0.07*	*0.01*
ES	*07/12/2018*	*22:00:00*	*34.50*	*15.19*	*0.09*	*0.03*
ES	*08/12/2018*	*02:00:00*	*34.77*	*14.44*	*0.15*	*0.00*
ES	*08/12/2018*	*06:00:00*	*35.43*	*13.93*	*0.07*	*0.00*
ES	*08/12/2018*	*10:00:00*	*36.06*	*13.44*	*0.13*	*0.05*
ES	*08/12/2018*	*14:00:00*	*36.64*	*12.99*	*0.11*	*0.04*
ES	*08/12/2018*	*18:00:00*	*37.22*	*12.54*	*0.21*	*0.09*
ES	*08/12/2018*	*22:00:00*	*37.84*	*12.04*	*0.16*	*0.00*
ES	*09/12/2018*	*02:00:00*	*38.57*	*11.46*	*0.10*	*0.02*
ES	*09/12/2018*	*06:00:00*	*39.30*	*10.88*	*0.15*	*0.02*
ES	*09/12/2018*	*10:00:00*	*39.98*	*10.32*	*0.21*	*0.01*
ES	*09/12/2018*	*14:00:00*	*40.70*	*9.73*	*0.20*	*0.02*
ES	*09/12/2018*	*18:00:00*	*41.41*	*9.14*	*0.16*	*0.01*
ES	*09/12/2018*	*22:00:00*	*42.10*	*8.56*	*0.14*	*0.00*
ES	*10/12/2018*	*02:00:00*	*42.82*	*7.95*	*0.20*	*0.02*
ES	*10/12/2018*	*06:00:00*	*43.00*	*7.79*	*0.15*	*0.01*
ES	*10/12/2018*	*10:00:00*	*43.31*	*7.53*	*0.30*	*0.12*
ES	*10/12/2018*	*14:00:00*	*44.00*	*6.92*	*0.29*	*0.03*
ES	*10/12/2018*	*18:00:00*	*44.73*	*6.29*	*0.39*	*0.04*
ES	*10/12/2018*	*22:00:00*	*45.30*	*5.78*	*0.37*	*0.03*
ES	*11/12/2018*	*02:00:00*	*45.80*	*5.33*	*0.30*	*0.00*
ES	*11/12/2018*	*06:00:00*	*46.29*	*4.90*	*0.31*	*0.01*
ES	*11/12/2018*	*10:00:00*	*46.77*	*4.45*	*0.33*	*0.03*
ES	*11/12/2018*	*14:00:00*	*47.32*	*3.94*	*0.32*	*0.02*
ES	*11/12/2018*	*18:00:00*	*47.86*	*3.44*	*0.30*	*0.01*
ES	*11/12/2018*	*22:00:00*	*48.35*	*2.99*	*0.31*	*0.02*
ES	*12/12/2018*	*02:00:00*	*48.82*	*2.54*	*0.31*	*0.02*
ES	*12/12/2018*	*06:00:00*	*49.24*	*2.14*	*0.25*	*0.00*
ES	*12/12/2018*	*10:00:00*	*49.69*	*1.71*	*0.31*	*0.01*
ES	*12/12/2018*	*14:00:00*	*50.09*	*1.31*	*0.29*	*0.05*
ES	*12/12/2018*	*18:00:00*	*50.60*	*0.82*	*0.24*	*0.01*
ES	*12/12/2018*	*22:00:00*	*51.12*	*0.30*	*0.33*	*0.03*
ES	*13/12/2018*	*02:00:00*	*51.73*	*0.00*	*0.23*	*0.04*
ES	*13/12/2018*	*06:00:00*	*52.50*	*0.00*	*0.22*	*0.02*
ES	*13/12/2018*	*10:00:00*	*53.30*	*0.00*	*0.24*	*0.03*
ES	*13/12/2018*	*14:00:00*	*54.00*	*0.00*	*0.32*	*0.01*
ES	*13/12/2018*	*18:00:00*	*54.48*	*0.00*	*0.31*	*0.01*
ES	*13/12/2018*	*22:00:00*	*55.28*	*-0.06*	*0.25*	*0.00*
ES	*14/12/2018*	*02:00:00*	*56.06*	*0.00*	*0.21*	*0.02*
ES	*14/12/2018*	*06:00:00*	*56.89*	*0.00*	*0.30*	*0.01*
ES	*14/12/2018*	*10:00:00*	*57.70*	*-0.01*	*0.32*	*0.02*
ES	*14/12/2018*	*14:00:00*	*58.52*	*0.00*	*0.30*	*0.00*
ES	*14/12/2018*	*18:00:00*	*59.35*	*-0.01*	*0.29*	*0.00*
ES	*14/12/2018*	*22:00:00*	*59.83*	*-0.01*	*0.34*	*0.00*
ES	*15/12/2018*	*02:00:00*	*60.38*	*-0.05*	*0.32*	*0.01*

ES	15/12/2018	06:00:00	61.10	0.00	0.28	0.00
ES	15/12/2018	10:00:00	61.58	-0.01	0.29	0.04
ES	15/12/2018	14:00:00	62.30	0.00	0.28	0.02
ES	15/12/2018	18:00:00	62.93	0.00	0.27	0.00
ES	15/12/2018	22:00:00	63.43	-0.02	0.28	0.02
ES	16/12/2018	02:00:00	63.94	0.00	0.25	0.00
ES	16/12/2018	06:00:00	64.50	0.00	0.22	0.02
ES	16/12/2018	10:00:00	65.40	-0.04	0.20	0.01
ES	16/12/2018	14:00:00	66.20	-0.01	0.24	0.01
ES	16/12/2018	18:00:00	66.99	0.00	0.22	0.02
ES	16/12/2018	22:00:00	67.92	-0.05	0.27	0.01
ES	17/12/2018	02:00:00	68.81	0.02	0.27	0.03
LS	27/2/2019	10:00:00	-70.26	-2.72	0.16	0.00
LS	27/2/2019	14:00:00	-69.99	-3.82	0.18	0.01
LS	27/2/2019	18:00:00	-69.64	-3.76	0.17	0.01
LS	27/2/2019	22:00:00	-68.84	-3.76	0.20	0.01
LS	28/2/2019	02:00:00	-67.99	-2.96	0.30	0.01
LS	28/2/2019	06:00:00	-67.04	-2.32	0.20	0.01
LS	28/2/2019	10:00:00	-65.04	-1.44	0.24	0.02
LS	28/2/2019	14:00:00	-64.94	-0.66	0.16	0.02
LS	28/2/2019	18:00:00	-63.90	0.00	0.19	0.00
LS	28/2/2019	22:00:00	-62.82	0.00	0.16	0.02
LS	1/3/2019	02:00:00	-62.18	0.00	0.18	0.00
LS	1/3/2019	06:00:00	-61.27	0.00	0.31	0.01
LS	1/3/2019	10:00:00	-60.16	-0.01	0.39	0.05
LS	1/3/2019	14:00:00	-60.01	-0.86	0.28	0.02
LS	1/3/2019	18:00:00	-59.97	-3.11	0.31	0.01
LS	1/3/2019	22:00:00	-59.88	-5.24	0.26	0.02
LS	2/3/2019	02:00:00	-59.86	-7.35	0.36	0.02
LS	2/3/2019	06:00:00	-59.79	-9.49	0.34	0.02
LS	2/3/2019	10:00:00	-59.73	-11.58	0.34	0.00
LS	2/3/2019	14:00:00	-59.74	-13.69	0.35	0.02
LS	2/3/2019	18:00:00	-59.70	-15.84	0.18	0.02
LS	2/3/2019	22:00:00	-59.64	-17.96	0.39	0.02
LS	3/3/2019	02:00:00	-59.62	-20.08	0.25	0.01
LS	3/3/2019	06:00:00	-59.58	-22.17	0.27	0.01
LS	3/3/2019	10:00:00	-59.54	-24.33	0.29	0.02
LS	3/3/2019	14:00:00	-59.48	-26.34	0.30	0.01
LS	3/3/2019	22:00:00	-59.00	-28.37	0.33	0.02
LS	4/3/2019	02:00:00	-58.32	-29.59	0.40	0.02
LS	4/3/2019	06:00:00	-57.62	-30.83	0.30	0.03
LS	4/3/2019	10:00:00	-56.99	-31.92	0.31	0.02
LS	4/3/2019	14:00:00	-56.48	-32.78	0.28	0.02
LS	4/3/2019	18:00:00	-55.96	-33.65	0.31	0.01
LS	4/3/2019	22:00:00	-55.43	-34.53	0.27	0.03
LS	5/3/2019	02:00:00	-54.89	-35.39	0.24	0.01

LS	5/3/2019	22:00:00	-54.18	-35.46	0.34	0.02
LS	6/3/2019	02:00:00	-54.17	-33.59	0.31	0.02
LS	6/3/2019	06:00:00	-54.16	-31.71	0.37	0.00
LS	6/3/2019	10:00:00	-54.15	-29.83	0.38	0.01
LS	6/3/2019	14:00:00	-54.11	-27.89	0.24	0.01
LS	6/3/2019	18:00:00	-54.12	-26.02	0.28	0.00
LS	6/3/2019	22:00:00	-54.01	-24.19	0.32	0.02
LS	7/3/2019	02:00:00	-54.12	-22.83	0.32	0.04
LS	7/3/2019	06:00:00	-54.12	-21.44	0.23	0.01
LS	7/3/2019	10:00:00	-54.09	-19.56	0.28	0.03
LS	7/3/2019	14:00:00	-54.09	-17.59	0.29	0.01
LS	7/3/2019	18:00:00	-54.09	-15.65	0.30	0.03
LS	7/3/2019	22:00:00	-54.09	-13.72	0.21	0.01
LS	8/3/2019	02:00:00	-54.06	-12.25	0.21	0.04
LS	8/3/2019	06:00:00	-54.06	-10.90	0.29	0.05
LS	8/3/2019	10:00:00	-54.04	-8.79	0.31	0.04
LS	8/3/2019	14:00:00	-54.04	-7.01	0.49	0.02
LS	8/3/2019	18:00:00	-54.03	-5.09	0.27	0.04
LS	8/3/2019	22:00:00	-54.02	-3.46	0.42	0.01
LS	9/3/2019	02:00:00	-54.01	-1.82	0.32	0.01
LS	9/3/2019	06:00:00	-54.00	-0.16	0.35	0.01
LS	9/3/2019	10:00:00	-53.56	-0.02	0.36	0.03
LS	9/3/2019	14:00:00	-52.78	0.00	0.26	0.01
LS	9/3/2019	18:00:00	-52.05	0.00	0.24	0.02
LS	9/3/2019	22:00:00	-51.45	0.00	0.32	0.09
LS	10/3/2019	02:00:00	-51.01	-0.22	0.27	0.03
LS	10/3/2019	06:00:00	-50.68	0.08	0.22	0.01
LS	10/3/2019	10:00:00	-50.40	1.43	0.32	0.08
LS	10/3/2019	14:00:00	-50.15	2.85	0.25	0.02
LS	10/3/2019	18:00:00	-49.61	3.95	0.32	0.03
LS	10/3/2019	22:00:00	-48.95	4.90	0.26	0.01
LS	11/3/2019	02:00:00	-48.23	5.82	0.36	0.09
LS	11/3/2019	06:00:00	-47.56	6.75	0.30	0.01
LS	11/3/2019	10:00:00	-46.76	7.00	0.27	0.01
LS	11/3/2019	14:00:00	-45.87	7.06	0.34	0.04
LS	11/3/2019	18:00:00	-44.99	7.36	0.28	0.01
LS	11/3/2019	22:00:00	-44.13	7.36	0.33	
LS	12/3/2019	00:00:00	-43.79	7.69	0.00	0.05
LS	12/3/2019	04:00:00	-43.18	7.81	0.15	0.04
LS	12/3/2019	08:00:00	-42.89	7.01	0.22	0.00
LS	12/3/2019	12:00:00	-42.12	8.01	0.25	0.11
LS	12/3/2019	16:00:00	-41.34	9.28	0.10	0.00
LS	12/3/2019	20:00:00	-40.54	9.94	0.02	0.01
LS	13/3/2019	00:00:00	-39.72	10.60	0.05	0.07
LS	13/3/2019	04:00:00	-38.89	11.25	0.01	0.01
LS	13/3/2019	08:00:00	-38.06	11.92	0.01	0.01

LS	*13/3/2019*	*12:00:00*	*-37.24*	*12.56*	*0.01*	*0.01*
LS	*13/3/2019*	*16:00:00*	*-36.42*	*13.19*	*0.02*	*0.01*
LS	*13/3/2019*	*20:00:00*	*-35.58*	*13.83*	*0.01*	*0.01*
LS	*14/3/2019*	*00:00:00*	*-34.92*	*14.32*	*0.00*	*0.00*
LS	*14/3/2019*	*04:00:00*	*-34.50*	*14.87*	*0.01*	*0.01*

Lines 219 and 222 – The authors should state the calculated p-values, and not just state that the values are or aren't significant. Moreover, are p-values really appropriate for this data set? I encourage the authors to think about this article: https://link.springer.com/article/10.1007/s10654-016-0149-3. Perhaps a simple comparison of the data would be more insightful than a "significant/not significant" binary outlook.

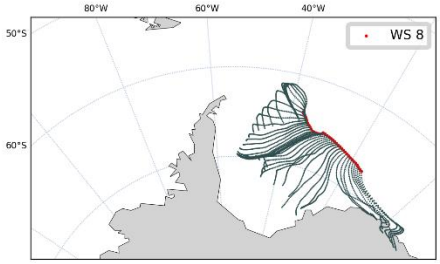
As per the reviewer's suggestion, we will include the calculated p-values in the text as opposed to simply stating whether values are or aren't significant. The conclusions drawn about the difference in the $\delta^{18}O$ data in the Weddell Sea vs. the latitudinal transect are primarily based on the spatial comparison (i.e., Figs 1B and 3). The statistical significance was added as a supporting note. We agree with the reviewer that relying on the significance of p-values to drive comparisons is not always valid, but still feel that it is relevant to keep the information in the manuscript with a more precise p-value provided.

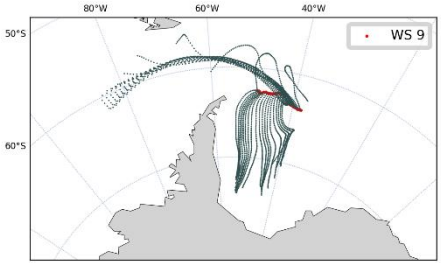
Figure 1 – the points in this figure are rather coarse, and it might be easier to see the points and error bars with thinner lines for both.

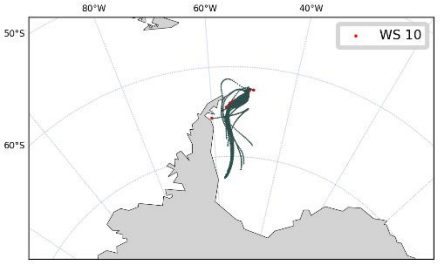
The linewidth of the symbols and error bars will be reduced by half to make the points clearer.

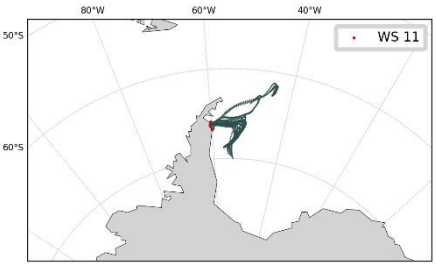
Figure 2 - It might make it somewhat more complicated, but it would be helpful to, rather than colour all the back trajectory lines the same tone of grey, to have them coloured by the date of the sample, even if it were done using an ombre (monotone) effect. In particular, this would be helpful for the Weddell Sea legs, because there is a significant amount of overlap in the back trajectories. Alternatively, individual or grouped back trajectories could be shown in a Supplemental Figure, to complement the complete regional back-trajectory version that is in this figure. Also, "AMBTs" doesn't need an apostrophe in the figure caption (and in the caption for Figure 3.)

The apostrophe in AMBTs will be removed from the figure captions of Figures 2 and 3.
We agree with the reviewer that there is significant overlap in AMBTs from the Weddell Sea, which makes it difficult to see exactly where the air from each separate filter deployment originated from. This is much clearer in the latitudinal early and later summer cruise transects as the filter deployments are spaced out. For interested readers we will include a supplementary figure that shows the AMBTs for every filter deployment from the Weddell Sea. This additional figure is shown below (Figure SX).


80°W
60°W
40°W
50°S
60°S
WS 8


80°W
60°W
40°W
50°S
60°S
WS 9


80°W
60°W
40°W
50°S
60°S
WS 10


80°W
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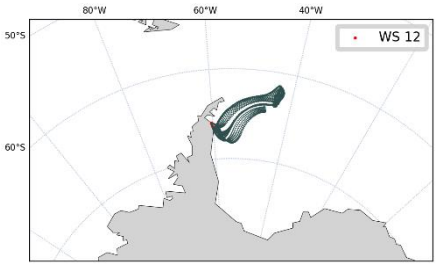
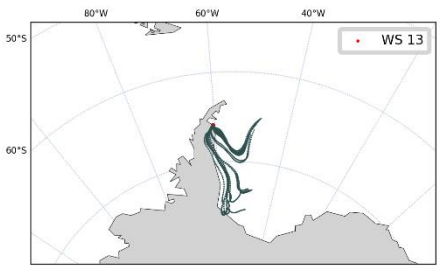
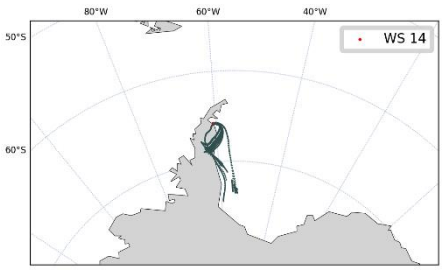
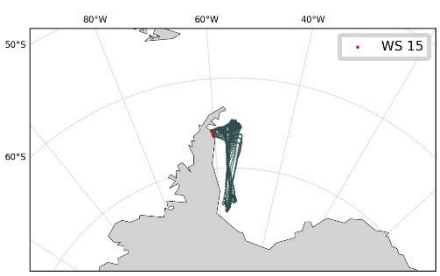
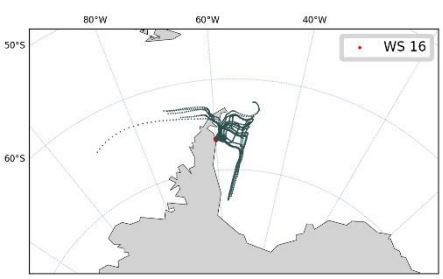
WS 11


80°W
60°W
40°W
50°S
60°S
WS 12


80°W
60°W
40°W
50°S
60°S
WS 13


80°W
60°W
40°W
50°S
60°S
WS 14


80°W
60°W
40°W
50°S
60°S
WS 15


80°W
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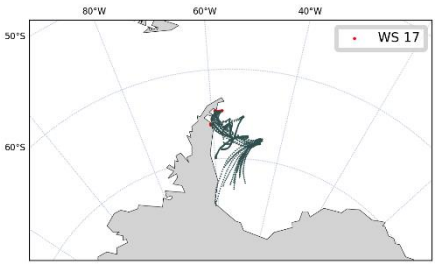
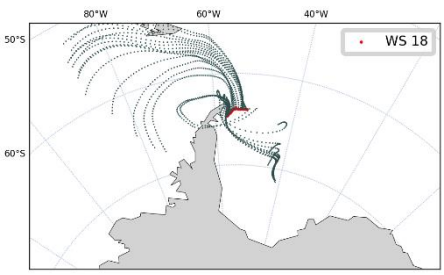
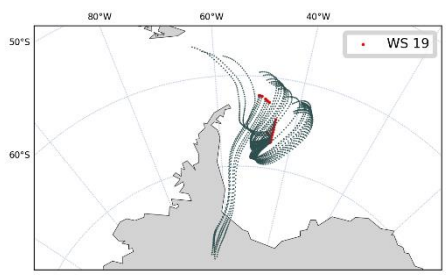
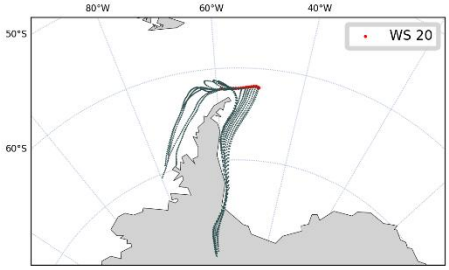
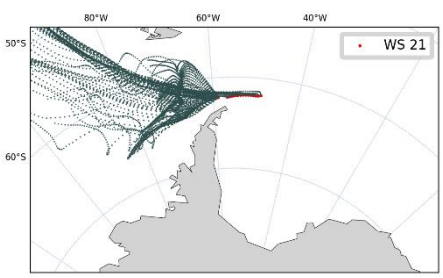
40°W
50°S
60°S
WS 16

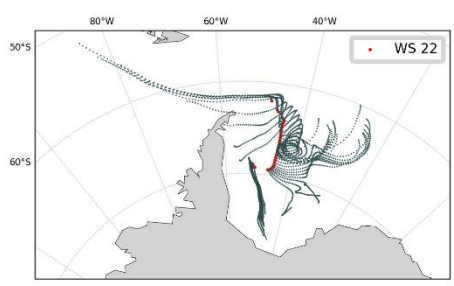

80°W
60°W
40°W
50°S
60°S
WS 17

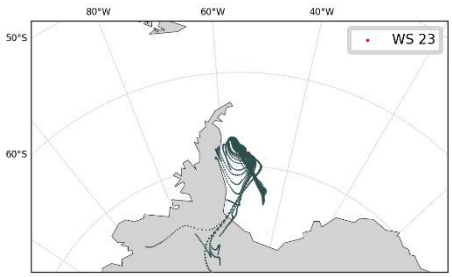

80°W
60°W
40°W
50°S
60°S
WS 18

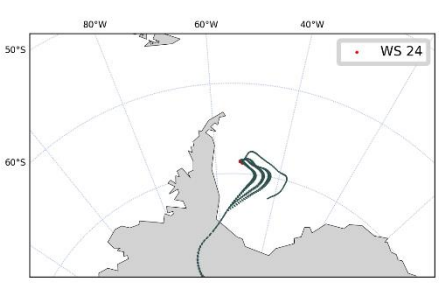

80°W
60°W
40°W
50°S
60°S
WS 19

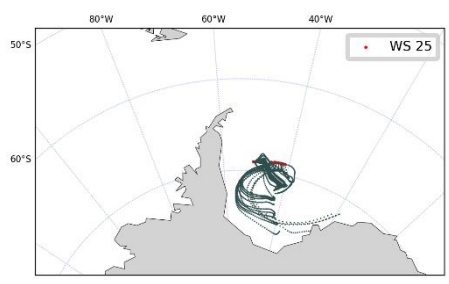

80°W
60°W
40°W
50°S
60°S
WS 20


80°W
60°W
40°W
50°S
60°S
WS 21


80°W
60°W
40°W
50°S
60°S
WS 22


80°W
60°W
40°W
50°S
60°S
WS 23


80°W
60°W
40°W
50°S
60°S
WS 24


80°W
60°W
40°W
50°S
60°S
WS 25

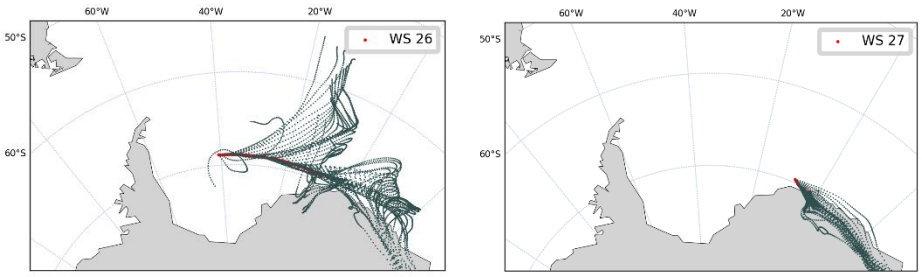


Figure SX. Maps depicting 72-hour AMBTs computed each hour for every filter deployment (dark grey dots) during the Weddell Sea leg of the research voyage. Each subplot represents one filter deployment, and the cruise leg ID is shown (Tables S1 and S3) in the legend on the upper right-hand side of each subplot. The red dots highlight the ships path.

Figure 4 - the back trajectory lines are again quite thick – if possible, it might be better to show the back trajectory lines with a slightly finer point size. Also, in the caption, lines 304-305 – "AMBTs" x2. Line 306 – "sea ice" (ice is a noun), and in line 307 "sea-ice concentration" (sea-ice is an adjective.) As well, "AMSR2 ASI programme" needs to be defined, either here or in the main text. Finally, "the white region represents the location…" of the sea ice identified by the AMSR2 ASI programme in the Antarctic study region, not "at the southernmost extent of each transect".

The apostrophe will be removed from both AMBTs instances.
"Sea ice" will be used everywhere in the caption when it is a noun, and "sea-ice" will be used when it is an adjective. The caption will be edited to reflect that the white shows the location of sea ice determined from satellite derived sea-ice concentration data obtained from the passive microwave sensor, AMSR2. AMSR2 will be described in the figure caption as "Advanced Microwave Scanning Radiometer 2, (Spreen et al., 2008)". The trajectory lines will also be plotted with a finer point to make them more visible.

Line 310 – many of the transects have two shades of orange, indicating that they originated in one region, and then continued through a different region before being sampled. Were these samples treated differently in your statistics than the ones that were modeled to be entirely in one region throughout the entire 72 hours? This needs to be clarified.

The AMBTs of all the samples originated from at least 2 zones, therefore their $\delta^{15}N\text{-}NO_3^-$ signature is derived from at least 2 different NO_x sources in all cases. There were no samples that were assumed to be entirely in one region throughout the entire 72 hours.
For each filter deployment (i.e., each sample), a 72-hour AMBT was run every hour. Each 72-hour AMBT for the whole filter deployment was taken into consideration when determining how much time was spent in each zone. This is clarified in lines 352-356 where the AMBTs are used along with the source NOx $\delta^{15}N$ values to derive the estimated alkyl nitrate influenced signal. The lack of clarity is perhaps due to the citation of figure 4 in line 310. We have amended the text so figure 2 is cited in line 310, which is more useful for evaluating the sentence "At the northern extent of our transects, the low-latitude aerosol samples, defined as those originating from anywhere north of 43°S in early summer and 41°S in late summer (Figure 2), had the highest average $\delta^{15}N\text{-}NO_3^-$ signature (-4.9 ±

1.5‰; n = 5).” As a result, the current Figure 4 (which will now be figure 5) will not be introduced until line 349 when the discussion of the use of AMBT to assist with determining the regions that contributed to the signal for each sample. Here we will also amend the text to clarify what the colours of the AMBTs in current Figure 4 represent. The text to be included is as follows: “We split the latitudinal transect into three regions, each characterised by the dominance of a different natural source of NO_3^- i.e., lightning NO_x at the low-latitudes (Figure 5 light orange), oceanic $RONO_2$ emissions at the mid-latitudes (Figure 5 dark orange) and snowpack emissions at the high-latitudes (Figure 5 red).”

In addition we have edited figures 2 and 3 to include the location of the sea ice.

Lines 318-319 – “end of the low-latitude zone” – this is still somewhat ambiguous.

The start of the mid-latitude zone which is also the end of the low latitudes zone was identified as the latitude at which non-zero sea surface nitrite concentrations begin to become prominent. We will correct this by explicitly stating these latitudes as 43°S and 41°S in early and late summer, respectively to make it clearer. In addition, we will delete the clause “i.e., end of the low latitude zone”. The sentence will now read:” The beginning of the mid-latitude zone (i.e., 43°S and 41°S in early and late summer, respectively) was defined by the presence of non-zero sea surface nitrite concentrations in early and late summer (Figure 4).”

Lines 363-370 – While I certainly appreciate the methodology being used here, there should be a discussion about the uncertainties inherent from putting so much of the analysis on the accuracy of the back trajectories. There needs to be a discussion about the reliability and uncertainties in the HYSPLIT AMBTs, and the resultant uncertainties in the calculated isotopic impacts on δ15N of aerosol nitrate.

We agree with the reviewer that a discussion of the uncertainty/reliability of HYSPLIT in producing AMBTs is useful considering the extent to which we rely on AMBT information to estimate the isotopic signature of aerosol nitrate derived from oceanic alkyl nitrates.

We are aware that HYSPLIT has limitations owing to multiple factors including the spatial and temporal resolution of the meteorological data used to force the model. The longer the back trajectory is, the less spatial coherency is observed, due to the propagation of error along back trajectories (Sinclair, et al., 2013). This can impact the accuracy of the location of each AMBT computed. According to literature the spatial uncertainty is estimated to be 15% to 30% of the travel distance because of errors in wind fields (Sarchilli et al., 2011).

Nevertheless, HYSPLIT is a frequently used tool for assessing air mass origin in the Southern Hemisphere and over Antarctica and is used in all three of the most cited papers for comparison purposes within this manuscript (Morin et al., 2008; Walters et al., 2019; Shi et al., 2021).

Perhaps most importantly with respect to this study, knowing the location of each AMBT with exact precision is not necessary given that we are operating over such large spatial scales. For example, if air masses originate from anywhere north of 43° S they are considered to be influenced by predominantly lightning NO_x. Variation in the path of an AMBT within a zone will not change the percent contribution of the dominant NO_x source in that zone to NO_3^-, or by extension the $\delta^{15}N$ value that we estimate for NO_3^- derived from $RONO_2$ emissions. The accuracy of AMBTs is perhaps more influential at the high latitudes where we assess contact with sea/continental ice. Even then, if we

simply assumed a threshold latitude and suggested that every AMBT located south of 70º is predominantly influenced by snow NO_x emissions, this would not change our results significantly (the $\delta^{15}N\text{-}NO_3^-$ originating from $RONO_2$ emissions in this case would be -23.5‰ as opposed to -22‰). However, the northern edge of the sea ice is not uniform with longitude, thus we used the AMBT locations that corresponded with locations of > 50% sea ice coverage in an attempt to be more accurate.

Furthermore, our AMBTs are short (72 hours), which alleviates some of the uncertainty caused by the propagation of error along back trajectories, as compared to studies with AMBTs of >5 days. We will add the following to the text to clarify the potential uncertainty associated with the AMBTs, and that this uncertainty is unlikely to influence the estimated $\delta^{15}N$ end member associated with alkyl nitrate emissions.

Text to be included at line 367: "Using this approach to estimate the $\delta^{15}N\text{-}NO_3^-$ from oceanic $RONO_2$ emissions relies heavily on AMBTs generated using HYSPLIT. While HYSPLIT is a frequently used tool for assessing air mass origin in the Southern Hemisphere and over Antarctica (Morin et al., 2008; Walters et al., 2019; Shi et al., 2021), it is important to note that a spatial uncertainty of 15% to 30% of the trajectory path distance can be expected (Sarchilli et al., 2011). AMBTs also become increasingly uncertain the further back in time they are used (Sinclair, et al., 2013). Some of this uncertainty is alleviated by the fact that the AMBTs generated here are relatively short. Additionally, the spatial scale of the low-, mid- and high-latitude zones is large, such that some variation in sample AMBTs will not significantly alter the samples dominant NO_3^- source."

Technical comments

Title – The period at the end of the title is unnecessary

The period at the end of the title will be removed.

Line 31 – here and throughout the paper, per the EGU style guide, use "and" instead of &, both in in-text citations and in the reference list. Similarly, per the style guide, for Figure panel labels, use lower case letters, i.e., (a), (b), etc. Also, "Coordinates need a degree sign and a space when naming the direction (e.g. 30° N, 25° E)", and "Common abbreviations to be applied: hour as h (not hr), kilometre as km, metre as m". Also, Figure captions should be numbered "Figure 1…", not "Fig. 1…", and Figures, Equations, and Sections should be referred to as "Fig. #, "Eq. (#)", and "Sect. #" when not at the beginning of a sentence. Likewise, reactions should be referenced in the text in parentheses: e.g., (R10).

We thank the reviewer for this technical guidance and will be sure to follow the EGU style guide by converting all "&" to "and" in both in-text citations and the reference list. We will also change figure panel labels to be in lower case letters. We will edit the co-ordinates to include a degree sign and a space. Lastly we will edit all figure captions to be numbered using "Figure X" as opposed to "Fig. X" and all reactions in the text will be correctly referenced using parentheses.

Line 31 – "Earth's"

This will be edited.

Line 38 – probably out to put "Southern Ocean (SO)" here, so later references to SO are defined.

"Southern Ocean (SO)" will be included.

Line 39 and throughout – references with "et al., YYYY" should not have a comma following the first author's last name.

Any case of a comma following the first authors last name will be removed in references with "et al., YYYY"

Line 53 – "(Jones et al., 2000, 2001)."

This will be edited as per the reviewer's suggestion.

Line 80 – remove the word "both" (three things are listed, so "both" doesn't make sense".)

The word "both" will be removed.

Line 86 – "R" should be italicized.

R will be italicized.

Line 94 – "(Berhanu et al., 2014, 2015)".

This will be edited as per the reviewer's suggestion.

Line 179 – Probably ideal to use the same notation for pooled standard deviation here and in Table S2, either sp (with subscripted p), or SDp (with a subscripted p.)

We will correct this by using same notation for pooled standard deviation here and in Table S2.

Line 190 – "BÓ§hlke et al."

This will be edited as per the reviewer's suggestion.

Line 198 – "ship's"

This will be edited as per the reviewer's suggestion.

Line 201 – This should probably be section 2.3, not 2.6. Also, the numbering notation of the section notation should be consistent throughout the manuscript for each type of heading: 1), 2), etc., or 1.1 Secondary Heading, 2.2 Another Secondary Heading, etc.

We will change this to refer to section 2.3 as opposed to 2.6 and ensure that our numbering notation for sections is consistent.

Line 207 (and Line 699) – I believe it is https://

This will be edited

Line 214 – "high latitudes"

This will be edited

Line 228 (Table 1) – N2 should have a subscripted "2".
The subscript "2" will be included.

Lines 228 (Table 1) and 230 (Figure 1 caption) – The convention for the standard notation, as you have on Line 87 and Table S2, is "VSMOW", not "V-SMOW".

The convention "VSMOW" will be used.

Line 241 – “Our observations reveal…” would be sufficient.

This will be edited.

Line 247 – “AMBTs”

This will be edited.

Lines 265-266 – pptv and ppbv should be defined.

We now only refer to ppbv in the text, and will define this as parts per billion by volume.

Line 287 – “high latitudes”
This will be edited.

Lines 323, 324, 360 – “AMBTs”
This will be edited.

Line 329 – “Dahl and Saltzman, 2008;”.
This will be edited.

Line 342 – “NOx” should have a subscripted x.

This subscript x will be included.

Line 351 – There should be a comma before “i.e.,”
A comma will be added before “i.e.,”

Lines 379-380 – recommend italicizing “*f*”, here and later.

We will italicize “f” here and elsewhere.

Line 383 and 384 – add a comma prior to i.e.
A comma will be added before “i.e.,”

Line 404 – “hypothesize” (or hypothesise for regional spelling consistency)
This will be edited.

Line 418 – HCl should have a lower case L.

A lower case “l” will be included here.
Line 449 – “AMBTs”, and the light blue lines aren’t dashed.
This will be edited.
Line 454 – “ for 15 January 2019”.
This will be edited.
Line 478 – “At the mid-latitudes, peroxy…” and “while in the”
Line 507 – “J.G.” (for consistency with other referenced (co-)authors.)
This will be edited.
Line 514 – use https://doi.org... formatting throughout the references. Also, here and throughout the reference list, per the EGU ACP guide, Journal Abbreviations should be used.
https://doi.org formatting will be used throughout the reference list and journal abbreviations will be used.

Line 527 – “Journal of Geophysical Research Letters” should be “J. Geophys. Res. - Atmos.”, and “Ocean” should be capitalized.

This will be edited.

Line 545 – C1 and C4 should have 1 and 4 subscripted. Also, “Letters” on the next line should also be “J. Geophys. Res. - Atmos.”
This will be edited.

Line 550 – also JGR-A, not GRL. And the DOI link is https://doi.org/10.1029/1999JD900238.
This will be edited.

Line 573 – “Saltzman, S. E.”
This will be edited.

Line 575 – “Craig”, not “Graig”; this article is the ACP version, so remove “Discussions” from line 577.
This will be edited and “Discussions” will be removed.

Line 579 – DOI citation: https://doi.org/10.1023/A:1009738715891
This will be included.
Line 580 – This should be “Elliott, E. M.” Also, in “United States,” (there is a space and a period where there should be a comma… likewise, in several other references, there is a period instead of a comma following the article title.)
This will be edited.

Line 587 – “nitrogen”
This will be edited.

Line 599 – there is a rogue comma detached from “Meteorology”.
This will be edited.

Line 601 – “… Research: Atmospheres”, DOI: https://doi.org/10.1029/93JD00874
This will be included.
Line 606 – “aerosol”
This will be edited.

Line 612 – “Atmospheric” is spelled incorrectly (although it should be “Atmos. …” and this is not a discussion paper, so remove “Discussions” from line 613.
“Discussions” will be removed and Atmospheric abbreviated.
Line 614 – if this is a book, it should have more details.
The full details will be included.

Line 619 – there is a space missing in “… vapor at…”, Maido should be “Maïdo”, and the DOI citation is: https://doi.org/10.1002/2017JD026791
This reference will be corrected and DOI included.
Line 628 - DOI citation: https://doi.org/10.1029/1999RG000078
This DOI will be included.

Line 630 – DOI citation: https://doi.org/10.1016/0016-7037(57)90021-2
This DOI will be included.

Line 643 – “Peroxyacetyl”
This will be edited.

Line 649 – The date for this citation (https://doi.org/10.1029/97JD02075) is in the wrong location.
The location of the date will be edited.
Line 655 – in the reference, it is spelled "Oxidized". And "Atmos." is missing from the Journal title.
This will be edited.

Line 660 – "formation"
This will be edited.

Line 674 – "Lee, H.-M., …" – the H is missing a period.
A period will be included.

Line 684 – "Atmos." is missing.
"Atmos." Will be included.

Line 687 – "Müller"
This will be edited.

Line 691 – "NOx" should have a subscripted x.
The x will be subscripted.

Line 705 – remove "Discussions."
Discussions will be removed.

Line 715 – "Galanter, M. and…"
This will be edited.

Line 727 – "Atmos." is missing.
"Atmos." Will be included.

Line 747 – "Atmos." is missing.
"Atmos." Will be included.

Line 754 – "Comparisons" is spelled incorrectly, and it should be "Atmospheric Environment" (no 's'), but of course, "Atmos. Environ."

Spelling will be corrected, and the abbreviated journal name will be used.

Line 754 – "NOx" should have a subscripted x.
The x will be subscripted.