Referee: 2

We thank Reviewer#2 for detailed comments and constructive suggestions, which have greatly improved the manuscript. Below, we provide a point-by-point response to the comments. Referee comments are in black, and the responses are in blue.

Sun et al. reported measurements of organic nitrogen (ON) across the global marine atmospheric boundary layer. They observed that ON/TN ratios in polar regions are significantly higher than in other regions, attributing this difference to marine biogenic emissions. Furthermore, in sea ice-covered areas of Antarctica, elevated ON concentrations were detected, with sea-ice-associated ecosystems proposed as the primary driver for enhanced ON production. Overall, this manuscript presents the first global-scale measurements of ON and provides measurement-based ON/TN ratios for the marine boundary layer, thereby offering a very valuable dataset and novel insights into atmospheric ON.

Response: We thank the reviewer for the positive assessment of our work. We have carefully addressed all comments and revised the manuscript accordingly to improve methodological clarity and interpretation.

Comment 1: One major concern is the potential lack of methodological details. Specifically, how did the authors calculate sea ice concentrations for individual samples? While remote sensing data are mentioned for illustrating spatiotemporal distributions of sea ice in the Southern Ocean, the types of remote sensing data employed are not specified, indicating a need for further clarification. Also see my comments below.

Response: Thank you for raising this important point. Sea ice concentrations (SIC) for individual samples were calculated using satellite-derived daily gridded passive microwave SIC products. For regional-scale visualization of sea-ice extent (SIE) and SIC variability, we used the Sea Ice Index (Version 3) distributed by NSIDC (Fetterer et al., 2017), derived from passive-microwave observations (SSM/I – SSMIS). For sample-level calculations, SIC values along trajectories were obtained from the AMSR2 daily gridded SIC product (University of Bremen, ASI algorithm; Version 5.4). For each trajectory endpoint, SIC was extracted by collocating the endpoint latitude/longitude and date with the daily SIC grid. We then calculated sample-level SIC as the mean SIC of all endpoints classified as sea-ice-covered.

We updated Section 2.7 and revised the manuscript to ensure clarity, as follows:

"*In this study, remote sensing data are utilized to illustrate the spatiotemporal distribution of sea ice concentrations (SICs) in the Southern Ocean. For regional-scale visualization of sea-ice extent (SIE) and SIC variability, we used the Sea Ice Index (Version 3) distributed by the National Snow and Ice Data Center*

(NSIDC) (Fetterer et al., 2017), which is derived from passive-microwave observations from DMSP SSM/I and SSMIS sensors (Cavalieri et al., 1997).

Sea-ice concentrations used here are derived from daily gridded passive-microwave SIC products, which provide all-weather coverage and are widely used for polar sea-ice monitoring. The SIC of each sample is calculated using the following formula:

$$SIC=\frac{\sum_{i=1}^{Ns} SIC_i}{Ns} \quad (7)$$

where SIC_i represents the average sea ice density at the endpoint of the specified track. Ns represents the total number of trajectory endpoints located on the sea ice area. For each trajectory endpoint, the SIC value was extracted by collocating the endpoint latitude/longitude and the corresponding day with the daily SIC grid; the sample-level SIC_i was then calculated as the mean SIC across all sea-ice-covered endpoints (Ns). Sea ice concentration data are from the AMSR2 dataset (Version 5.4. University of Bremen, Germany. Index of /amsr2/asi_daygrid_swath/s3125)."

References:

Cavalieri, D. J., Gloersen, P., Parkinson, C. L., Comiso, J. C., and Zwally, H. J.: Observed Hemispheric Asymmetry in Global Sea Ice Changes, Science, 278, 1104–1106, https://doi.org/10.1126/science.278.5340.1104, 1997.

Fetterer, F., Knowles, K., Meier, W. N., Savoie, M. and Windnagel, A. K.: Sea Ice Index. (G02135, Version 3). [Data Set]. Boulder, Colorado USA. National Snow and Ice Data Center. https://doi.org/10.7265/N5K072F8, 2017.

Comment 2: L54-55: It may be more appropriate to attribute the elevated ON to sea-ice-associated ecosystems rather than sea ice dynamics.

Response: Thank you for raising this important point. We agree that it is more precise to attribute the elevated ON to sea-ice-associated ecosystems rather than broadly to "sea-ice dynamics". We revised the text accordingly.

"*Near Antarctic, ON concentrations and ON/TN ratios were distinctly elevated in sea-ice-influenced air masses, highlighting the role of sea-ice-associated ecosystems as a likely driver of enhanced ON production and emissions.*"

Comment 3: L98-99: In open oceans, do previous studies suggest a strong correlation between ocean primary productivity and ON or WSON? In polar regions, sea ice variability directly affects primary productivity and consequently ON levels, implying a potential linkage to sea ice dynamics.

Response: Previous studies have indeed reported correlations between ocean primary productivity and atmospheric WSON, including in open-ocean environments (e.g., Altieri et al., 2016). We have revised the text to clarify that such relationships have been observed. For polar environments, we now more explicitly describe sea ice variability as a plausible indirect driver of ON by modulating primary productivity

and related emissions.
We revised the main text, as follows:

"*Yet open-ocean and polar regions, where sea ice variability can strongly modulate primary productivity and thus potentially influence ON emissions, remain sparsely observed, limiting constraints on potential sea ice linked controls on ON, especially for high latitudes (Altieri et al., 2016; Matsumoto et al., 2022).*"

Reference:

Altieri, K. E., Fawcett, S. E., Peters, A. J., Sigman, D. M., and Hastings, M. G.: Marine biogenic source of atmospheric organic nitrogen in the subtropical North Atlantic, Proc. Natl. Acad. Sci. U.S.A., 113, 925–930, https://doi.org/10.1073/pnas.1516847113, 2016.

Matsumoto, K., Kobayashi, H., Hara, K., Ishino, S., and Hayashi, M.: Water-soluble organic nitrogen in fine aerosols over the Southern Ocean, Atmos. Environ., 287, 119287, https://doi.org/10.1016/j.atmosenv.2022.119287, 2022.

Comment 4: L104: Perhaps both Antarctic and Arctic campaigns should be referenced, as indicated in Section 2.1 on sample collection.

Response: Thank you for the suggestion. The text has been revised to explicitly reference both Antarctic and Arctic campaigns, consistent with the description in Section 2.1, as follows:

"*To address these gaps, we measured aerosol ON and IN using samples collected during three Chinese Arctic and Antarctic research expedition campaigns, spanning ~160° of latitude from the Arctic to Antarctica.*"

Comment 5: L194: Should the title of Section 2.5 be revised to "Potential Source Contribution Function analysis" for consistency with the main text?

Response: Thank you for the suggestion. The section title has been revised to "Potential Source Contribution Function (PSCF) analysis" for consistency with the main text.

Comment 6: L222: Why was a 20 km radius selected for the AEC index calculation? Additionally, why were pressures below 850 hPa assigned a Chl-a value of zero? These aspects require clarification.

Response: The 20 km radius was selected to balance spatial representativeness with the native resolution of the satellite Chl-a product, ensuring sufficient valid pixels while minimizing spatial smoothing. Chl-a was set to zero when trajectory endpoints were located over land, sea ice, or at pressures below 850 hPa because air masses at these altitudes are generally decoupled from local ocean surface biological activity. We added this content in Section 2.6 of the article as follows:

"*For each trajectory point, Chl-a concentrations (Chla$_i$) were obtained from satellite remote sensing products (Aqua-MODIS, OCI algorithm; 8-day composite, 4 km × 4 km resolution; https://oceancolor.gsfc.nasa.gov/l3/) within a 20-km radius to* *reduce the influence of missing/cloud-contaminated pixels and pixel-scale noise, while remaining small enough to preserve local marine biological variability relevant to each trajectory point. The 20 km radius approach has been widely adopted in previous studies to mitigate the uncertainty of trajectory endpoints and ensure robust matching with satellite data coverage in previous research (Park et al., 2018; Zhou et al., 2021, 2023). Trajectory endpoints over Antarctica, sea-ice-covered areas, or at pressures < 850 hPa were assigned Chl-a = 0 because air masses at these altitudes are generally decoupled from local ocean surface biological activity (Zhou et al., 2023).* "

Reference:

Park, K., Lee, K., Kim, T., Yoon, Y. J., Jang, E., Jang, S., Lee, B., and Hermansen, O.: Atmospheric DMS in the Arctic Ocean and Its Relation to Phytoplankton Biomass, Global Biogeochemical Cycles, 32, 351–359, https://doi.org/10.1002/2017GB005805, 2018.

Zhou, S., Chen, Y., Paytan, A., Li, H., Wang, F., Zhu, Y., Yang, T., Zhang, Y., and Zhang, R.: Non-Marine Sources Contribute to Aerosol Methanesulfonate Over Coastal Seas, JGR Atmospheres, 126, e2021JD034960, https://doi.org/10.1029/2021JD034960, 2021.

Zhou, S., Chen, Y., Wang, F., Bao, Y., Ding, X., and Xu, Z.: Assessing the Intensity of Marine Biogenic Influence on the Lower Atmosphere: An Insight into the Distribution of Marine Biogenic Aerosols over the Eastern China Seas, Environ. Sci. Technol., 57, 12741–12751, https://doi.org/10.1021/acs.est.3c04382, 2023.

Comment 7: L282-284: This raises an important point: could the previously underestimated percentage result from the exclusion of WION? If so, can atmospheric ON fractions, WION and WSON, be quantified or estimated?

Response: The previously underestimated ON/TN percentage could partly result from the exclusion of WION. In principle, if both total ON and WSON are measured for the same samples, WION can be calculated by difference (WION = total ON – WSON), and the relative fraction can be expressed accordingly. However, at this stage we are unable to robustly quantify atmospheric ON fractions (WION vs. WSON) for our dataset. First, the very limited aerosol mass in remote/clean environments makes conventional WSON determination (aqueous extraction) difficult and often sample-volume constrained. Second, we are concerned that uncertainties associated with the traditional WSON extraction/measurement approach may propagate into large errors when estimating WION by difference, potentially biasing the inferred fractions. The quantification of WION and WSON partitioning may be an important topic for future work. We appreciate the reviewer's suggestion, which highlights an important methodological issue and will be carefully considered in the future studies.

Comment 8: L299-304: Might secondary marine sources involve organic species from biological activities that undergo atmospheric oxidation to form ON? Beyond reactions with acidic species, are there alternative production pathways in the marine boundary layer?

Response: Thank you for this insightful suggestion. We agree that the description of secondary marine ON sources require expansion. In the revised manuscript, we have expanded this section to include the oxidative pathways of biogenic volatile organic compounds (BVOCs).
Specifically, we now acknowledge that marine BVOCs (e.g., isoprene and monoterpenes) emitted by phytoplankton undergo atmospheric oxidation via OH radicals (daytime) and NO_3 radicals (nighttime) to form secondary ON (Fisher et al., 2016). The NO_3-initiated oxidation in the marine boundary layer is a particularly efficient pathway for producing multifunctional organic nitrates (Ng et al., 2017). Furthermore, we have noted that small-molecule alkyl nitrates can also be produced via photochemical reactions of dissolved organic matter (DOM) in the sea surface microlayer and then released into the atmosphere (Chuck et al., 2002).

The revised text is as follows:

"*ON in the MABL primarily originates from two main source pathways: marine emissions and long-distance continental transport. Marine sources include primary ON, predominantly associated with sea-spray particles enriched in biological material from the ocean surface microlayer, and secondary ON. The latter not only derives from marine precursors such as alkylamines that react with acidic species (Altieri et al., 2016; Facchini et al., 2008), but also significantly involves the atmospheric oxidation of marine-derived biogenic volatile organic compounds (BVOCs). Specifically, isoprene and monoterpenes emitted from the ocean can react with hydroxyl (OH) or nitrate radicals (NO_3) to form secondary organic nitrates (Fisher et al., 2016; Ng et al., 2017). Additionally, direct sea-to-air emissions of light alkyl nitrates produced photochemically in the surface water contribute to the MABL ON pool (Chuck et al., 2002).*"

References:

Altieri, K. E., Fawcett, S. E., Peters, A. J., Sigman, D. M., and Hastings, M. G.: Marine biogenic source of atmospheric organic nitrogen in the subtropical North Atlantic, Proc. Natl. Acad. Sci. U.S.A., 113, 925–930, https://doi.org/10.1073/pnas.1516847113, 2016.

Chuck, A. L., Turner, S. M., and Liss, P. S.: Direct Evidence for a Marine Source of C1 and C2 Alkyl Nitrates, Science, 297, 1151–1154, https://doi.org/10.1126/science.1073896, 2002.

Facchini, M. C., Decesari, S., Rinaldi, M., Carbone, C., Finessi, E., Mircea, M., Fuzzi, S., Moretti, F., Tagliavini, E., Ceburnis, D., and O'Dowd, C. D.: Important Source of Marine Secondary Organic Aerosol from Biogenic Amines, Environ. Sci. Technol., 42, 9116–9121, https://doi.org/10.1021/es8018385, 2008.

Fisher, J. A., Jacob, D. J., Travis, K. R., Kim, P. S., Marais, E. A., Chan Miller, C., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., Mao, J., Wennberg, P. O., Crounse, J. D., Teng, A. P., Nguyen, T. B., St. Clair, J. M., Cohen, R. C., Romer, P., Nault, B. A., Wooldridge, P. J., Jimenez, J. L., Campuzano-Jost, P., Day, D.

A., Hu, W., Shepson, P. B., Xiong, F., Blake, D. R., Goldstein, A. H., Misztal, P. K., Hanisco, T. F., Wolfe, G. M., Ryerson, T. B., Wisthaler, A., and Mikoviny, T.: Organic nitrate chemistry and its implications for nitrogen budgets in an isoprene- and monoterpene-rich atmosphere: constraints from aircraft (SEAC4 RS) and ground-based (SOAS) observations in the Southeast US, Atmos. Chem. Phys., 16, 5969–5991, https://doi.org/10.5194/acp-16-5969-2016, 2016.

Ng, N. L., Brown, S. S., Archibald, A. T., Atlas, E., Cohen, R. C., Crowley, J. N., Day, D. A., Donahue, N. M., Fry, J. L., Fuchs, H., Griffin, R. J., Guzman, M. I., Herrmann, H., Hodzic, A., Iinuma, Y., Jimenez, J. L., Kiendler-Scharr, A., Lee, B. H., Luecken, D. J., Mao, J., McLaren, R., Mutzel, A., Osthoff, H. D., Ouyang, B., Picquet-Varrault, B., Platt, U., Pye, H. O. T., Rudich, Y., Schwantes, R. H., Shiraiwa, M., Stutz, J., Thornton, J. A., Tilgner, A., Williams, B. J., and Zaveri, R. A.: Nitrate radicals and biogenic volatile organic compounds: oxidation, mechanisms, and organic aerosol, Atmos. Chem. Phys., 17, 2103–2162, https://doi.org/10.5194/acp-17-2103-2017, 2017.

Comment 9: Regarding "continental sources," does this refer to ON formed over continents and subsequently transported to oceanic regions?

Response: In this manuscript, "continental sources" refers to the aggregate influence associated with air masses originating from (or transported over) continental regions. This includes: (1) ON formed directly over land and transported offshore, and (2) ON formed in situ during transport from continental precursors (e.g., anthropogenic NO_x and VOCs). Because our dataset does not allow us to distinguish these two sub-pathways, we use the term "continental sources" to represent their aggregate contribution.

We have revised the text to acknowledge the existence of both mechanisms, as follows:

"*Continental sources involve the long-range transport of organic emissions—including combustion byproducts, soil- and vegetation-derived compounds, and biomass burning aerosols. It is important to note that these continental inputs include both ON formed directly over land and ON produced from continental precursors during transport (Duce et al., 2008; Li et al., 2025). This transport can significantly influence remote ocean regions (Cape et al., 2011; Jickells et al., 2013).*"

References:

Cape, J. N., Cornell, S. E., Jickells, T. D., and Nemitz, E.: Organic nitrogen in the atmosphere — Where does it come from? A review of sources and methods, Atmos. Res., 102, 30–48, https://doi.org/10.1016/j.atmosres.2011.07.009, 2011.

Duce, R. A., LaRoche, J., Altieri, K., Arrigo, K. R., Baker, A. R., Capone, D. G., Cornell, S., Dentener, F., Galloway, J., Ganeshram, R. S., Geider, R. J., Jickells, T., Kuypers, M. M., Langlois, R., Liss, P. S., Liu, S. M., Middelburg, J. J., Moore, C. M., Nickovic, S., Oschlies, A., Pedersen, T., Prospero, J., Schlitzer, R.,

Seitzinger, S., Sorensen, L. L., Uematsu, M., Ulloa, O., Voss, M., Ward, B., and Zamora, L.: Impacts of Atmospheric Anthropogenic Nitrogen on the Open Ocean, Science, 320, 893–897, https://doi.org/10.1126/science.1150369, 2008.

Jickells, T., Baker, A. R., Cape, J. N., Cornell, S. E., and Nemitz, E.: The cycling of organic nitrogen through the atmosphere, Phil. Trans. R. Soc. B, 368, 20130115, https://doi.org/10.1098/rstb.2013.0115, 2013.

Li, Y., Fu, T.-M., Yu, J. Z., Zhang, A., Yu, X., Ye, J., Zhu, L., Shen, H., Wang, C., Yang, X., Tao, S., Chen, Q., Li, Y., Li, L., Che, H., and Heald, C. L.: Nitrogen dominates global atmospheric organic aerosol absorption, Science, 387, 989–995, https://doi.org/10.1126/science.adr4473, 2025.

Comment 10: L306: How was it determined that air masses spent 22.6% of their 5-day trajectories over continental areas? Is a single trajectory track used per sample? Methodological details appear insufficient, aligning with general concerns.

Response: Thank you for pointing this out. We agree additional methodological detail is needed. The percentage (e.g., 22.6%) represents the time-weighted residence-time fraction of trajectory endpoints over continental grid cells within the 5-day backward trajectories. For the full dataset, we used a representative midpoint for each 48 h sample and generated trajectories at 6 h intervals; residence-time fractions were then computed by classifying endpoints (open ocean/sea ice/continent) and applying an exponential time-weighting to account for dispersion and removal during transport. For the Antarctic sea-ice analysis subset, we additionally applied a moving-track ensemble approach (48 hourly positions per sample) and computed the same residence-time metrics using the ensemble of trajectories.

We revised Section 2.4 to describe this nested strategy and to define the residence-time calculation.

"To study air mass origins, air mass backward trajectories have been calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectories (HY-SPLIT) model with meteorological fields from the National Oceanic and Atmospheric Administration (NOAA) air resources laboratory GDAS database. Five-day backward trajectories were calculated in order to reveal the history of the air masses arriving at the sampling site (Stein et al., 2015). Each trajectory originated at the vessel's real-time position with an arrival height of 20 m, capturing boundary layer transport while minimizing local ship influence. Air mass backward trajectories were simulated using the HY-SPLIT model with meteorological fields from the NOAA GDAS database to reveal the transport history of air masses arriving at the vessel (Stein et al., 2015). Given that the ship was continuously moving and each sample integrates air masses over approximately 2–4 degrees of latitude, we applied a nested strategy to account for spatiotemporal variability. For the initial characterization of the entire dataset, a representative sampling location was defined for each sample using the average latitude and longitude of its start and end positions, with backward trajectories simulated at 6-h intervals anchored to this midpoint to identify dominant air-mass

categories (Fig. 3). Subsequently, to precisely investigate the influence of sea ice on ON in the Southern Ocean and Antarctic marginal regions (Section 4.2), a targeted high-resolution analysis was performed on this subset of samples. For each Antarctic sample, the actual cruise track was equally divided into 48 points corresponding to the hourly intervals of the 48-h sampling period, and a 120-h backward trajectory was calculated for each of these 48 coordinates (Fig. S3a and b).

To determine whether the backward trajectories of the MABL samples were mainly influenced by the open ocean, sea-ice-covered regions, or the continental area, we calculated the time-weighted residence-time ratios of air masses over sea ice (R_S), open ocean (R_O), and the continental area (R_C) using the following equation:

$$R_S(R_O\ or\ R_C)=\frac{\sum_{i=1}^{N_S(N_O\ or\ C)}\times e^{-(\frac{t_i}{120})}}{\sum_{i=1}^{N_{total}}\times e^{-(\frac{t_i}{120})}} \qquad (4)$$

where N_{total} denotes the total number of trajectory endpoints; N_S N_O and N_C represent the numbers of endpoints located over sea ice, the open ocean, and the Antarctic ice sheet, respectively. t_i is the backward-trajectory time (in hours), and $t_i/120$ is a time-weighting factor (Zhou et al., 2021). This factor accounts for air-mass dispersion during transport and aerosol removal by particle deposition; therefore, regions associated with longer trajectory times exert weaker influences on the sampling site, whereas nearby regions exert stronger influences. Accordingly, higher values of R_S, R_O and R_C indicate greater influences from sea ice, the open ocean, and the Antarctic ice sheet, respectively. ”

References:

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bull. Am. Meteorol. Soc., 96, 2059–2077, https://doi.org/10.1175/BAMS-D-14-00110.1, 2015.

Zhou S, Chen Y, Paytan A, et al. Non-Marine Sources Contribute to Aerosol Methanesulfonate Over Coastal Seas [J]. Journal of Geophysical Research: Atmospheres, 2021, 126: e2021JD034960.

Comment 12: Figure 4: Note that EC is not an ionic species.
Response: Thank you for noting this. The figure caption and associated text have been corrected.

Comment 13: L364-368: Employing a longer time interval in backward trajectory analysis might reveal air masses traversing continents. As demonstrated, EC detected in remote oceans confirms long-range transport from continental sources. Similarly, fine ON particles could be transported to the remote Southern Ocean. The absence of correlation between ON and EC or nssCa^{2+} may indicate that ON is not predominantly of continental origin.
Response: We agree that extending the trajectory duration would provide a more

comprehensive history of air mass origins and is particularly useful for identifying the transport of long-lived species like EC. In this study, we selected a 5-day trajectory interval primarily to align with the typical atmospheric residence time of ON (Textor et al., 2006; Cape et al., 2011). This window allows us to focus on the sources most likely to influence the observed concentrations.

Our chemical tracer data supports the reviewer's insight regarding the distinction between long-range transport and local production. While continental air masses may indeed reach the Southern Ocean (bringing trace EC), the distinct lack of correlation between ON and continental tracers ($nssCa^{2+}$, EC) suggests that the continental contribution to the ON pool is minimal. This decoupling reinforces our conclusion that the observed ON is predominantly driven by local marine processes.

We have revised the text to reflect this understanding, as follows:

"*In the SO region, ON concentrations were the lowest among all regions (mean = 12.0 ± 7.1 ng* m^{-3}*), yet the ON/TN ratio was relatively high (27.8 ± 11.0%). Back-trajectory analysis indicates that air masses predominantly originated from the open ocean and Antarctic continent (Fig. 3d), with minimal anthropogenic influence. ON here exhibited a significant positive correlation with* Na^+ *(Fig. 4;* $r = 0.43$, $p < 0.01$*), but no significant relationships with* $nssK^+$, $nssCa^{2+}$ *or EC. While long-range transport events may deliver stable continental tracers like EC to this remote region, the lack of correlation between ON and these markers suggests that continental inputs are not the primary driver of ON variability. This pattern, combined with the association with* Na^+ *suggests that primary sea-salt emissions are an important pathway for ON in the SO atmosphere (Matsumoto et al., 2022), likely through the incorporation of marine-derived organic matter into sea-spray aerosols. Meanwhile, the absence of associations with terrestrial tracers further supports the notion that ON in this remote region is controlled primarily by natural marine processes rather than continental or anthropogenic sources (Altieri et al., 2016).*"

References:

Altieri, K. E., Fawcett, S. E., Peters, A. J., Sigman, D. M., and Hastings, M. G.: Marine biogenic source of atmospheric organic nitrogen in the subtropical North Atlantic, Proc. Natl. Acad. Sci. U.S.A., 113, 925–930, https://doi.org/10.1073/pnas.1516847113, 2016.

Cape, J. N., Cornell, S. E., Jickells, T. D., and Nemitz, E.: Organic nitrogen in the atmosphere — Where does it come from? A review of sources and methods, Atmos. Res., 102, 30–48, https://doi.org/10.1016/j.atmosres.2011.07.009, 2011.

Matsumoto, K., Kobayashi, H., Hara, K., Ishino, S., and Hayashi, M.: Water-soluble organic nitrogen in fine aerosols over the Southern Ocean, Atmos. Environ., 287, 119287, https://doi.org/10.1016/j.atmosenv.2022.119287, 2022.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkev,

A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 2006.

Comment 14: L377-382: Previous studies have reported high primary productivity at sea ice edges.

Response: We agree. The sea ice edge is indeed a hotspot for biological activity, characterized by intense phytoplankton blooms and high primary productivity triggered by increased light availability and water column stratification during ice retreat.

In the revised manuscript, we have explicitly incorporated this point:

"*Sea-ice and open-ocean environments create distinct conditions for the production and emission of ON. While sea ice restricts direct air–sea exchange, it hosts specialized microbial communities and accumulates organic matter within brine channels. During melt and ice-edge retreat, this organic material is released into waters characterized by high primary productivity (Arrigo et al., 2008). This biological intensification enriches the surface microlayer and supplies precursors for aerosolization via sea spray and secondary formation (Dall'Osto et al., 2017; DeMott et al., 2016; Galgani et al., 2016; Wilson et al., 2015).*"

References:

Arrigo, K. R., Van Dijken, G. L., and Bushinsky, S.: Primary production in the Southern Ocean, 1997–2006, J. Geophys. Res., 113, 2007JC004551, https://doi.org/10.1029/2007JC004551, 2008.

Dall'Osto, M., Ovadnevaite, J., Paglione, M., Beddows, D. C. S., Ceburnis, D., Cree, C., Cortés, P., Zamanillo, M., Nunes, S. O., Pérez, G. L., Ortega-Retuerta, E., Emelianov, M., Vaqué, D., Marrasé, C., Estrada, M., Sala, M. M., Vidal, M., Fitzsimons, M. F., Beale, R., Airs, R., Rinaldi, M., Decesari, S., Cristina Facchini, M., Harrison, R. M., O'Dowd, C., and Simó, R.: Antarctic sea ice region as a source of biogenic organic nitrogen in aerosols, Sci Rep, 7, 6047, https://doi.org/10.1038/s41598-017-06188-x, 2017.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F., and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, Proc. Natl. Acad. Sci. U.S.A., 113, 5797–5803, https://doi.org/10.1073/pnas.1514034112, 2016.

Galgani, L., Piontek, J., and Engel, A.: Biopolymers form a gelatinous microlayer at

the air-sea interface when Arctic sea ice melts, Sci Rep, 6, 29465, https://doi.org/10.1038/srep29465, 2016.

Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. A., Nájera, J. J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Temprado, J. V., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. A., and Murray, B. J.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525, 234–238, https://doi.org/10.1038/nature14986, 2015.

Comment 15: L389: "sea-ice–linked" should be corrected to "sea-ice–associated" for consistency.
Response: Thank you for the suggestion. The wording has been corrected throughout the manuscript for consistency:

"*Multiple lines of evidence point to sea-ice–associated biological processes as the driver of these enhancements...*"

Comment 16: Figure 5 caption: The statement "owing to missing satellite data and the methodologies in Sections 2.6 and 2.7" is ambiguous.
Response: Thank you for pointing this out. We clarify that the sample size for the correlations was reduced for two reasons. First, satellite products for SIC and Chl-a are not always available along the trajectories (e.g., due to gaps/clouds). Second, following our data-screening criteria, SIC or Chl-a is considered valid for a sample only when at least 75% of the trajectory points have valid satellite values. We have revised the Figure 5 caption accordingly.

We revised the caption of Figure 5 as follows:

"*Figure 5. Comparison of measured ON concentrations (a) and ON/TN ratio (b) between SI and OO aerosol samples ("**" indicating $p < 0.01$). And correlations between SIC (c), AEC (d) and ON concentration in SI aerosol samples. The sample sizes are $n = 10$ for panel (c) and $n = 6$ for panel (d). These reduced sample sizes are due to unavailable satellite SIC/Chl-a data along the trajectories, and in this study SIC or Chl-a is used only when ≥75% of the trajectory points have valid satellite values (see Sections 2.6–2.7).*"

Comment 17: L456-460: If WION was substantially underestimated in prior studies, the current observations hold significant climate relevance, as WION may function as cloud condensation nuclei.
Response: We thank the reviewer for highlighting this critical implication. We fully agree that the underestimation of ON in prior studies (due to the omission of WION) has profound consequences for our understanding of aerosol-cloud interactions.

As the reviewer correctly points out, WION components—such as biological colloids and proteinaceous matter—can significantly influence cloud microphysics. They can act directly as cloud condensation nuclei (CCN) or enhance CCN activity by lowering surface tension (Ovadnevaite et al., 2011). Furthermore, insoluble organic particles are known to be efficient Ice Nucleating Particles (INP) in the marine atmosphere (Wilson et al., 2015).

In the revised manuscript, we have explicitly expanded this section:

> "*Comparison with prior WSON-only datasets suggests that earlier studies likely underestimated total ON—by approximately 40% in the Southern Ocean—due to omission of WION. Accounting for both soluble and insoluble phases is therefore essential for constraining nitrogen deposition to the oceans and for representing ON's roles in atmospheric processes. Specifically, given that WION may significantly influence cloud condensation nuclei activity and cloud droplet formation, overlooking this fraction could lead to substantial uncertainties in assessing the radiative forcing and climate effects of marine aerosols.*"

References:

Ovadnevaite, J., Ceburnis, D., Martucci, G., Bialek, J., Monahan, C., Rinaldi, M., Facchini, M. C., Berresheim, H., Worsnop, D. R., and O'Dowd, C.: Primary marine organic aerosol: A dichotomy of low hygroscopicity and high CCN activity: MARINE AEROSOL-CLOUD INTERACTIONS, Geophys. Res. Lett., 38, n/a-n/a, https://doi.org/10.1029/2011GL048869, 2011.

Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. A., Nájera, J. J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Temprado, J. V., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. A., and Murray, B. J.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525, 234–238, https://doi.org/10.1038/nature14986, 2015.

Comment 18: Please double-check the Supplementary material. In particular, the full name instead of abbreviation may be better in the Figure captions.

Response: Thank you for the reminder. The Supplementary material has been carefully checked, and abbreviations in figure captions have been replaced with full names where appropriate.

End of responses to the Referee.