Referee: 1

We are grateful to reviewer#1 for the detailed comments and constructive suggestions, which have greatly improved the manuscript. Below we provide a point-by-point response. Referee comments are in black, and our responses are in blue.

This manuscript presents the first direct measurements of aerosol organic nitrogen (ON) along a global marine atmospheric boundary layer transect, revealing clear latitudinal and regional patterns. The study is based on substantial and carefully conducted field campaigns and addresses an important knowledge gap in marine atmospheric nitrogen cycling. The results are potentially impactful for understanding ON sources and for improving atmospheric and climate model representations. I therefore recommend publication of this manuscript after the issues and questions raised below are adequately addressed.

Response: We appreciate the Reviewer's positive comments on our manuscript. Below, we give a point-by-point response to your comments and suggestions.

Special comments:

(1) Line 33. Please check the unit "ng mm^{-3}" is right?

Response: Thank you for pointing this out. It has now been corrected to "ng m^{-3}" in the abstract.

(2) Line 35. The variance of ON/TN ratio should also be mentioned

Response: We agree. In the revised manuscript, we have added the statistical variability of the ON/TN ratio to better characterize its spatial variability along the transect. This has been added to both the Abstract and the Results sections.

We revised the main text as follows:

Abstract: "*A significant latitudinal gradient was observed, with significantly higher ON concentrations in the Northern Hemisphere (83.3 ± 141.4 ng m*$^{-3}$*) than in the Southern Hemisphere (15.4 ± 12.4 ng m*$^{-3}$*). Regionally, coastal East Asia recorded the highest ON levels (164.6 ± 179.1 ng m*$^{-3}$*) but a lower ON/TN ratio (21.1 ± 7.9%), indicating strong terrestrial and anthropogenic influence. In contrast, the Arctic Ocean had lower ON concentrations (19.1 ± 19.0 ng m*$^{-3}$*) but the highest ON/TN ratio (38.6 ± 12.4%), suggesting dominant marine biogenic sources. The Southern Ocean showed the lowest ON concentration (12.0 ± 7.1 ng m*$^{-3}$*) yet a relatively high ON/TN ratio (27.8 ± 11.0%), also pointing to oceanic origins.*"

Results: "*The CEA region exhibited the highest ON concentrations (mean = 164.6 ng m*$^{-3}$*) but the lowest ON/TN ratio (mean = 21.1 ± 7.9%). In contrast, the SO region showed the lowest ON concentrations (mean = 12.0 ng m*$^{-3}$*; range: 1.78–32.3 ng m*$^{-3}$*)*

and higher ON/TN ratios (mean = 27.8 ± 11.0%). Notably, the AO region displayed the highest ON/TN ratios (mean = 38.6 ± 12.4%) despite relatively low ON concentrations (mean = 19.1 ng m^{-3}; range: 5.2–32.2 ng m^{-3}). The ON/TN ratio in SATO region (26.8 ± 10.0%) is similar to that of SO but with a lower ON concentration (mean = 23.4 ng m^{-3}; range: 5.7–70.1 ng m^{-3}), which is much lower than the CEA region, but higher than the high latitude two pole regions."

(3) Line 56-102. The narrative flow of the "Introduction" could be improved. For example, the first paragraph highlights the limitations of traditional ON analytical approaches but does not immediately introduce your solutions; and the discussion of ON sources in the second and fourth paragraphs is interrupted by the third paragraph on the geochemical significance of ON, which makes the overall structure somewhat difficult to follow.

Response: We thank the reviewer for this constructive suggestion. To improve the narrative flow, we restructured the Introduction. Specifically, we moved the key solution statement (simultaneous IN/ON measurement capturing both WSON and WION without subtraction) to the end of the first paragraph to immediately address the limitation raised. We also grouped all source-related discussion consecutively and moved the paragraph on the geochemical/climate significance after the source discussion so that the overall structure is easier to follow.

The revised Introduction is as follows:

"Marine atmospheric boundary layer (MABL) aerosol particles contain significant amounts of organic nitrogen (ON) and inorganic nitrogen (IN), both recognized as major components of atmospheric particulate matter (Li et al., 2023). ON may account for roughly 20–80% of total reactive nitrogen deposition to the surface ocean, implying a potentially large, yet uncertain, role in marine nitrogen cycling and climate (Altieri et al., 2016, 2021). However, ON remains poorly constrained due to analytical limitations (Baker et al., 2017). Previous studies focused on the water-soluble fraction of aerosol ON (WSON) inferred indirectly by subtraction IN from total nitrogen (TN) (ON = TN – IN), while the water-insoluble organic nitrogen (WION) fraction has been largely unquantified (Cornell, 1999; Mace et al., 2003). The subtraction approach is prone to errors and artifacts, especially when TN and IN concentrations are similar, leading to underestimation and large uncertainties in ON burdens and fluxes. To overcome these limitations, we employed a newly developed aerosol nitrogen analyzer based on thermal evolution and chemiluminescence detection to measure aerosol IN and ON simultaneously, eliminating subtraction-based method biases and capturing both WSON and WION (Yu et al., 2021).

Aerosol ON arises from diverse sources. Marine pathways include primary emissions via sea spray enriched with organic matter from the sea surface microlayer and secondary formation from marine precursors (e.g., alkylamines) reacting with acidic species (Facchini et al., 2008; Miyazaki et al., 2011a). Continental pathways include long-range transport of organic emissions from fossil fuel combustion, *biomass burning, soils, and vegetation (Cape et al., 2011; Jickells et al., 2013; Luo et al., 2018). Primary marine emissions inject large amounts of particulate matter annually, carrying organic carbon and nitrogen from plankton, bacteria, and surface films (Violaki et al., 2015a). Observations have shown that sea spray can carry substantial ON and that WION can dominate ocean-influenced aerosol ON (Miyazaki et al., 2011a).*

Despite this importance, ON sources over remote oceans remain debated. Some studies implicate continental transport (e.g., dust, anthropogenic emissions), whereas others point to direct sea spray emissions or secondary formation from marine-derived alkylamines (Altieri et al., 2016; Lesworth et al., 2010; Zamora et al., 2011). Correlations between ON and ocean biological proxies (e.g., chlorophyll-a) suggest in situ marine production, particularly during phytoplankton blooms (Altieri et al., 2016; Dall'Osto et al., 2019). 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 (Matsumoto et al., 2022). Around Antarctica in particular, the paucity of direct ON measurements—especially of WION—limits understanding of ON sources, seasonality, and impacts on high-latitude atmospheric chemistry.

ON affects climate and biogeochemistry by supplying bioavailable nitrogen, modifying cloud condensation nuclei and ice-nucleating particle populations, and contributing to aerosol light absorption. Hygroscopic ON compounds (e.g., amino acids, amines, sugars) enhance water uptake and cloud condensation nuclei (CCN) activity; some proteinaceous organics act as efficient ice nuclei (Alsante et al., 2024; Chan et al., 2005). Marine alkylamines can form salts with sulfuric acid, promoting new particle formation and growth, thereby linking ON to aerosol number and radiative forcing (Almeida et al., 2013; Brean et al., 2021). Nitrogen-containing chromophores (brown nitrogen) can dominate the absorptive properties of organic aerosol regionally and contribute substantially to global absorption by carbonaceous aerosol (Li et al., 2025).

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. Using this analyzer, this dataset enables evaluation of hemispheric and regional patterns, assessment of controlling factors (e.g., continental influence, marine biological activity), and explicit investigation of sea-ice–associated processes near Antarctica. The results provide observational constraints that can be used to refine the representation of nitrogen cycling and atmosphere–ocean interactions in climate and atmospheric chemistry models."

(4) Line 169-184. Given the very low ON concentrations in marine aerosol samples, it would be helpful for the authors to clarify the detection limit of the new method and whether it offers a clear advantage over traditional IC-based approaches? Additionally,

more information on the separation and discrimination between IN and ON would strengthen confidence in the measurements.

Response: The ON measurements in this study follow the thermal evolution–chemiluminescence method developed by Yu et al. (2021), which reports a method detection limit of 96 ng N (absolute mass on the analyzed filter aliquot). In practice, low ambient concentrations can be reliably measured by increasing the analyzed filter area. In this study, we typically used 4–6 cm^2 of filter per sample for analysis, which proportionally lowers the effective concentration detection limit and ensures reliable ON quantification.

Compared with traditional IC-based approaches, this method offers a clear advantage by enabling the simultaneous determination of IN and ON on the same filter aliquot, thereby avoiding the difference method (ON = TN − IN) and the associated large uncertainty propagation when TN and IN are similar in magnitude. IN/ON discrimination is based on their distinct thermal evolution behavior and the joint interpretation of C and N thermograms: ON is identified by co-evolving C and N signals across temperature steps, whereas IN is characterized by N-only evolution without a corresponding C signal. Overlapping features are further separated using multivariate curve resolution (MCR), as validated using laboratory standards, synthetic mixtures, and ambient aerosol samples in Yu et al. (2021).

We have added the method detection limit description, the effective detection limit used in this study, and a clearer explaination of the IN/ON separation and discrimination (C–N thermograms combined with MCR/PMF) to the Methods section, as follows:

"*Aerosol ON and IN were simultaneously measured using the recently developed Aerosol Nitrogen Analyzer system, which enables sensitive quantification directly from filter samples without pretreatment. Detailed descriptions of the method are provided in Yu et al (2021). Briefly, the method detection limit is 96 ng N. Because the detection limit scales inversely with the analyzed filter area, it can be readily lowered by analyzing a larger aliquot. In this study, 4–6* cm^2 *of filter material was typically analyzed for each sample, yielding a proportionally lower effective detection limit and ensuring stable and reliable quantification for low-concentration marine aerosol samples. Compared with traditional IC-based approaches, this analyzer provides a clear advantage by determining IN and ON simultaneously on the same filter aliquot, thereby avoiding the subtraction-based "difference method" (ON = TN – IN) and the associated uncertainty propagation when TN and IN are similar in magnitude.*

The analyzer integrates a thermal aerosol carbon analyzer and a chemiluminescence NOx analyzer. Aerosol samples collected on quartz fiber filters were thermally evolved under a programmed 6-step temperature protocol (150, 180, 300, 400, 500, and 800 °C) in a 1% O_2*/99% He carrier gas. The evolved materials were catalytically oxidized to* CO_2 *and nitrogen oxides (*NO_y*), with the C signal monitored via methanator-FID detection and the N signal recorded through chemiluminescence* *after converting NO_y to NO. The C signal assists in differentiating IN and ON components, as ON aerosols produce both C and N signals while the IN fraction only yields an N signal. The programmed thermal evolution facilitates separation of aerosol IN and ON due to their distinct thermal characteristics. Specifically, IN and ON discrimination is achieved by jointly interpreting the C and N thermograms: ON is identified by co-evolving C and N signals across the temperature steps, whereas IN is characterized by N-only evolution without a corresponding C signal. The separation of overlapping thermal features is further resolved using multivariate curve resolution (MCR), which deconvolves the mixed thermograms into source-like components based on their distinct thermal evolution patterns. Quantification of IN and ON is achieved through multivariate curve resolution (MCR) data treatment of the C and N thermograms using USEPA PMF (version 5.0).*"

Reference:

Yu, X., Li, Q., Ge, Y., Li, Y., Liao, K., Huang, X. H., Li, J., and Yu, J. Z.: Simultaneous Determination of Aerosol Inorganic and Organic Nitrogen by Thermal Evolution and Chemiluminescence Detection, Environ. Sci. Technol., 55, 11579–11589, https://doi.org/10.1021/acs.est.1c04876, 2021.

(5) Line 191. I didn't get it. As the ship was continuously moving and each sample integrates air masses over approximately 2–4 degrees of latitude, it is not entirely clear how the backward air-mass trajectories were constructed for an individual sample. The authors should clarify how the temporal and spatial variability during sampling was accounted for in the trajectory analysis.

Response: We agree that, because the ship was continuously moving and each sample integrates air masses over ~2–4° of latitude, the trajectory analysis must account for spatiotemporal variability during the 48 h sampling period. We therefore implemented a two-step trajectory analysis strategy. First, for the entire dataset, a standard characterization was conducted using a representative midpoint approach (the average of the start and end coordinates) with trajectories generated at 6-hour intervals. This provides a consistent, large-scale overview of the air-mass history for all samples. However, we recognize that this simplified approach may not capture the fine-scale heterogeneity required for investigating sea-ice influences.

Accordingly, for the Southern Ocean and Antarctic marginal regions (Section 4.2), we conducted a high-resolution, moving-track strategy. For each 48 h sample, we divided the cruise track into 48 points (hourly positions) and calculated a 120 h (5-day) backward trajectory for each point. This high-resolution ensemble approach explicitly accounts for the ship's continuous movement and the spatial integration of the samples, providing a robust basis for calculating parameters such as sea-ice residence time and diagnostics. We have clarified this in the revised Methods text:

“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 (5-day) backward trajectory was calculated for each of these 48 coordinates (Fig. S3a and b).”

Reference:

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.

(6) Line 222. Why 20-km radius was chosen?

Response: We selected a 20 km radius to balance robustness and spatial representativeness when matching satellite Chl-a to trajectory points. The Aqua-MODIS Level-3 Chl-a product used in this study has a spatial resolution of 4 km × 4 km; a 20-km radius typically includes >20 pixels, which reduces the influence of missing data (e.g., clouds) and local pixel-scale noise.

At the same time, this radius remains sufficiently small to preserve local marine biological varibility relevant to the air-mass residence area represented by each trajectory point. Similar spatial averaging scales have been commonly adopted in previous trajectory-based marine aerosol studies (Park et al., 2018; Zhou et al., 2021, 2023). We have added this rationale in Section 2.1:

“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.

(7) Line 245 and Figure5. Given the large variability in the dataset and limited data numbers, it would be advisable to assess the normality of the data in the Supporting Information to confirm whether the use of t-tests is statistically appropriate (or other statistical methods such as Mann-Whitney U test).

Response: We thank the reviewer for this valuable suggestion. Data normality was assessed using the Shapiro-Wilk test, and the results are provided in the Supporting Information (Table S1). Because several datasets deviated from normality; we used the non-parametric Mann-Whitney U tests. The statistical significance and overall conclusions are consistent with those obtained using t-tests, supporting the robustness of the results shown in Line 245 and Figure 5.

This point was clarified in the revised manuscript:

Section 3: “*Atmospheric ON concentrations exhibited significant hemispheric differences ($p < 0.001$; Mann–Whitney U test; Table S1), with values in the Northern Hemisphere (NH: 83.3 ± 141.4 ng m^{-3}, $N = 55$) being approximately five times higher than those in the Southern Hemisphere (SH: 15.4 ± 12.4 ng m^{-3}, $N = 37$).*”

Section 4.2: “*Along the Antarctic coast, we classified samples into two groups based on air-mass histories: open ocean (OO), influenced almost exclusively by open-ocean trajectories, and sea ice (SI), with air masses residing over sea ice for extended periods. SI samples exhibited significantly higher ON concentrations and ON/TN ratios than OO samples (; $p < 0.001$; Mann–Whitney U test; Fig. 5a, b).*”

Table S1. Normality assessment and non-parametric statistical tests for comparisons

Comparison	*Group A (n)*	*Group B (n)*	*Shapiro–Wilk p (A)*	*Shapiro–Wilk p (B)*	*Test applied*	*p-value*
ON (NH vs SH)	*51*	*40*	2.27×10^{-11}	1.02×10^{-6}	*Mann–Whitney U*	8.43×10^{-4}
ON (Sea-ice vs non–sea-ice)	*13*	*11*	*0.585*	7.58×10^{-4}	*Mann–Whitney U*	2.01×10^{-3}
ON/TN (Sea-ice vs non–sea-ice)	*13*	*11*	*0.0107*	*0.69*	*Mann–Whitney U*	9.01×10^{-4}

(8). Line 276. I am confused about how total nitrogen (TN) was determined using ion chromatography in previous studies (Table 1); could the authors clarify whether the reported WSON/TN ratios in the literature actually refer to WSON/WSTN instead?

Response: We appreciate this important comment. In the studies summarized in Table 1, “TN” was determined on aqueous extracts (e.g., via persulfate oxidation, UV oxidation, or TOC/TN analyzer methods), and therefore represents **water-soluble total nitrogen (WSTN)** rather than total aerosol nitrogen. Accordingly, the ON/TN ratios reported in the literature should be interpreted as WSON/WSTN rather than total aerosol nitrogen. This point has now been clarified in Table 1.

(9) Table1: I suggest to add your own dataset in Table1

Response: Thanks for this suggestion. We have now added our dataset to Table 1. This inclusion facilitates direct comparison between our global marine transect observations and previously reported studies.

The modified table is as follows:

Table 1. Measured concentrations of WSON from published reports in the marine atmospheric boundary layer.

Regions	Period	Locations	WSON (ng m^{-3})	Ratio to WSTN(%)	Methods for IN	Methods for WSTN	Reference
Southern Atlantic	2007.01-02	35° S–45° S	119 ± 163.8	-	IC	PO	(Violaki et al., 2015b)
the Northwest Pacific Ocean	2014.2015	25–40° N, 125–150° E	43.4–564.2	11–46	IC	PO	(Luo et al., 2018)
the coast of China the East China seas	2014.2015	30–40° N, 120–130° E	96.6–7238	6–48	IC	PO	(Luo et al., 2018)
Southern Ocean	2000.11	40.41° S, 144.41° E	50.4 ± 79.8	21	IC	UV	(Mace et al., 2003)
Oahu, Hawaii	1998.7.22-8.13	21.7º N, 157.8º W	46.2	31	IC	UV	(Cornell et al., 2001)
the southern margin of the East China Sea	2005-2006	25.09° N, 121.46° E	476 ± 756	24 ± 16	IC	UV with PO	(Chen and Chen, 2010)
the western North Pacific	2008.08.24-09.13	42.98° N,144.37° E –35.65° N, 139.77° E	130 ± 61 (10–260)	67 ± 15	IC	TOC/TN analyzer	(Miyazaki et al., 2011b)
Huaniao Island	2019	30.86° N, 122.67° E	30–2810	0.13–77	IC	TOC/TN analyzer	(Tian et al., 2023)
the northern tip of Japan	2010-2012	~45.2° N, ~141.2° E	77 ± 57	12.8 ± 15.2	IC	TOC/TN analyzer	(Matsumoto et al., 2017)
the Southern Ocean	2016-2020	38.8–69.0º S, 38.1–150.8º E	4.7	20	IC	TOC/TN analyzer	(Matsumoto et al., 2022)
the Subarctic Western North Pacific Ocean	2016.7.21-8.22	30–65º N, 130–160º W	1.62–205.8	9	IC	TOC/TN analyzer	(Jung et al., 2019)
Bermuda	2011	32.27° N, 64.87° W	105 ± 191.8	50.4 ± 18.9	nutrient analyzer	TN Analyzer	(Altieri et al., 2016)
the Arctic Ocean	2021.07-09	north of ~60° N	5.2–32.2	38.6 ± 12.4	N Analyzer	N Analyzer	this study
the Coastal East Asia	2021.07-09, 2019.10-11, 2023.10-2024.04	20–60° N	18.1–555.6	21.1 ± 7.9	N Analyzer	N Analyzer	this study
Athe Southeast Asia-Australia Tropical Ocean	2021.11, 2023.11	20° N–40° S	5.7–70.1	26.8 ± 10.0	N Analyzer	N Analyzer	this study
the Southern Ocean	2021.11-2022.03, 2019.11, 2023.11	south of ~ 40° S	1.8–32.3	27.8 ± 11.0	N Analyzer	N Analyzer	this study

*PO: the persulfate oxidation (PO) method

*UV: ultraviolet photo-oxidation

*TN analyzer: a total organic carbon (TOC) analyzer with a TN unit

*Nutrient analyzer: automated nutrient analyzer and standard colorimetric method

(10) Line 295 "4.1 Source identification of ON". As correlation alone does not allow one to distinguish between common emission sources and shared atmospheric transport or processing, I suggest that the authors either temper causal language (e.g., “dominated by”, “primarily controlled by”) or provide additional independent evidence to strengthen the source attribution.

Response: We agree that correlation alone cannot unambiguously separate common emission sources from shared transport and/or atmospheric processing. Our source interpretation is not based on correlations alone. We also consider (i) air-mass origin and residence-time diagnostics from backward trajectories (marine/continental/sea-ice/Antarctic influence), (ii) consistency with established source tracers (e.g., Na^+ for sea-salt/sea-spray influence, $nssCa^{2+}$ for crustal/dust inputs, $nssK^+$ for biomass burning, EC for combustion-related influence), (iii) contrasts between air-mass categories (e.g., continental-influenced vs. marine-only) supported by statistical tests, and (iv) marine biological proxies (AEC derived from Chl-a along trajectories). These complementary constraints help identify the most plausible dominant contributors to ON variability in each region.

Nevertheless, to avoid over-interpreting correlations, we revised Section 4.1 to temper causal wording (e.g., replacing “dominated by/primarily controlled by” with “suggests/consistent with/likely influenced by”) and to explicitly reference the multiple lines of evidence used.

The revised text is provided below:

“*A significant correlation between ON and the anthropogenic tracer EC ($r = 0.81$, $p < 0.01$; Fig. 4) indicates that fossil fuel combustion and biomass burning are important ON sources .*”

“ *Meanwhile, the absence of associations with terrestrial tracers further supports the notion that ON in this remote region is likely influenced more significantly by natural marine processes rather than continental or anthropogenic sources.*”

“*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.*”

(11) Line 327-335. The author said that the correlation between ON and nss-Ca^{2+} is good, but there is no correlation between ON and nss-K^+ or EC. Is this a contradiction? I suggest the authors further explain this inconsistency. From the perspective of air masses, the backward trajectories of SATO samples have few intersections with the continental region. Therefore, can nss-Ca^{2+} be regarded as a reliable indicator of continental transport?

Response: This is a very insightful question. We agree that the different behavior of $nssCa^{2+}$ versus $nssK^+$ and EC in the SATO region requires clarification. For the SATO samples, the air-mass origins can be divided into two types: (1) trajectories that traveled only over the ocean and (2) trajectories that passed over land (Fig. R1). Figure S1a shows a significant difference between these two groups. The ON concentrations are significantly higher in the land-influenced samples than in the ocean-only samples. Specifically, although some air masses passed over land (e.g., Southeast Asian islands and coastal Australia), these regions do not possess the same high-intensity emission profiles as the East Asian continent.

In this context, nssCa^{2+} serves as a tracer of mineral/dust from the continents, which will affect remote marine air via long-range transport in the free troposphere followed by subsidence and mixing into the marine boundary layer. Different from nssCa^{2+}, nssK^+ and EC are more closely associated with biomass burning/combustion emissions. The lack of ON–nssK^+ and ON–EC correlations suggests that combustion-related influence is limited and not the dominant driver of ON variability in this subset, whereas the ON–nssCa^{2+} relationship points to episodic mineral influence or other non-combustion terrestrial sources (e.g., soil organic matter associated with dust, terrestrial biogenic particles, or agricultural emissions). We have revised the text to clarify these points and to avoid treating nssCa^{2+} as an exclusive proxy for continental transport, as follows:

"*The SATO region exhibits intermediate level of ON concentrations (mean = 23.4 ± 18.0 ng* m^{-3}*), lower than those influenced by anthropogenic activities in CEA but higher than in polar regions. In this region, ON shows a significant positive correlation with nss*Ca^{2+} *(Fig. 4;* $r = 0.76$, $p < 0.01$*), suggesting that terrestrial mineral inputs (e.g., dust) influence ON levels, rather than purely marine sources. In addition, backward trajectory analysis showed that samples affected by continental air masses have significantly higher ON concentrations than those exposed solely to marine air (Fig. 3c), suggesting the influences of continental sources. However, ON does not exhibit significant correlations with nss*K^+ *or with EC (*$p > 0.05$*), indicating that combustion emissions may not be the dominant driver in this region. These findings suggest that the variability of ON in the SATO region results from a mixture of marine-terrestrial interactions, primarily modulated by episodic terrestrial mineral influence rather than continuous marine emissions. Notably, this region displays an elevated ON/TN ratio (Fig. 2), primarily due to its very low IN levels—approximately 85% lower than in the CEA region—which amplifies the relative contribution of ON within TN.*"

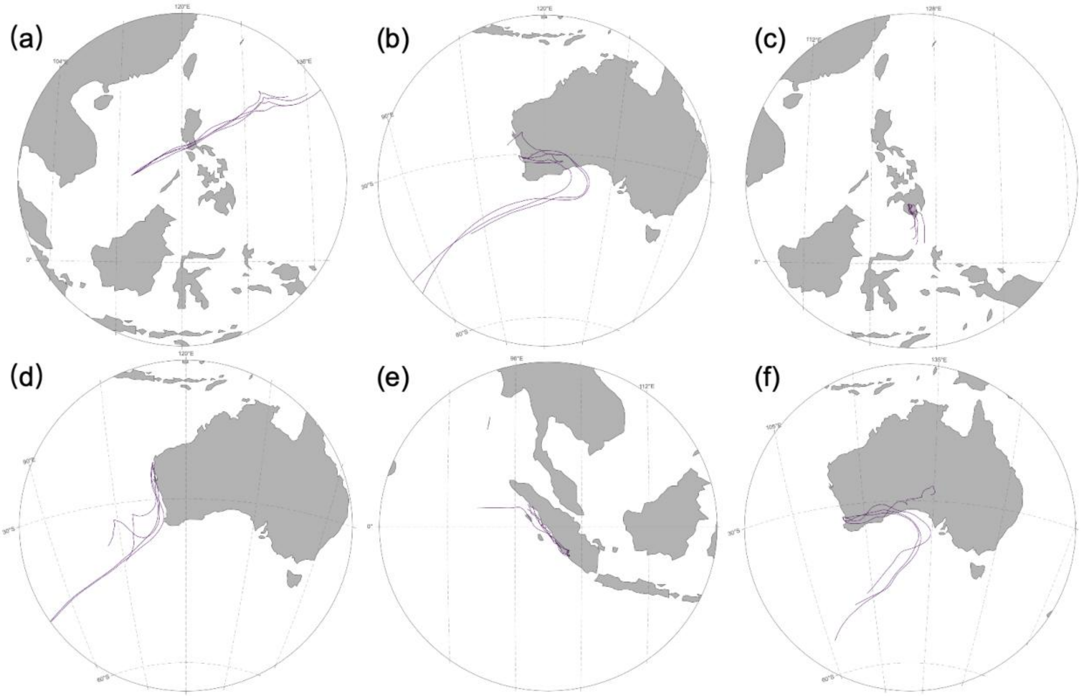

Figure R1. Analysis of backward trajectory for the samples collected in the Southeast Asia-Australia Tropical Ocean region on November 10, 2021 (a), November 13, 2021 (b), November 20, 2021 (c), November 10, 2023 (d), November 12, 2023(e) and November 14, 2023 (f).

End of responses to the Referee.