

## Reviewer#1

*This is an interesting article on providing data and results on the origins of WSON (and WSIN) in a coastal area in Thailand (Bangkok). Different approaches are used, chemical speciation, back-trajectories, source apportionment and remote sensing derived data.*

*The tasks are well implemented and interesting results are obtained. However, there is a need for a very relevant review before accepting this article to ACP. Some refers to the methodology, others to the need of additional data analyses and some about interpretation of results. See below general and specific comments.*

**Response:** We appreciate this careful and constructive assessment. The comments facilitated a more precise understanding of the original manuscript's shortcomings, particularly with regard to the discussion of source attribution, sampling limitations, and the level of confidence attached to marine-biogenic interpretations. The manuscript has undergone extensive revision in light of the aforementioned concerns.

### General

- 1. To obtain the relevance of specific contributions of a source to an atmospheric pollutant/component, one to several years of sampling are needed, but samples should be obtained constantly along the year to obtain representative annual data. If excluded periods and more intensively sampling is done in specific ones, the relevance of specific contributions can be not representative. Here you sampled more intensively in some, and avoided others.*

**Response:** Thank you for this important comment. Our sampling covered a full year, but not with uniform temporal coverage, especially during the rainy period. We have therefore clarified in the Methods that the dataset should be interpreted as a year-long observational record rather than as a uniformly sampled annual climatology, and we now state this limitation explicitly again in the Conclusions.

Revised text in the Method on page 5, lines 134–139:

“Because the sampling frequency varied among seasons and was reduced during the rainy period, this dataset does not represent a uniformly sampled annual climatology. Accordingly, the results are interpreted as observation-based estimates for the sampled annual cycle, and caution is needed when extending them to annual-scale representativeness.”

Revised text in the Conclusions on page 22, lines 624–630:

“However, these implications should be interpreted with caution because the present dataset was obtained with seasonally uneven sampling coverage, 24 h integrated filter sampling, and without isotopic constraints that could more directly distinguish marine-biogenic, shipping-related, and continental anthropogenic sources. Future work combining more temporally uniform observations with isotopic and molecular-level characterization is needed to further strengthen source attribution.”

2. *Should one use special sampling protocols, denuders, filter packs,..... to avoid the loss of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and ON, or the artifact formation? How this may influence your results?*

**Response:** Denuder- or filter-pack-based sampling would indeed be preferable when the aim is to minimize gas–particle artifacts in  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and particulate organic nitrogen measurements. Because our study relied on integrated filter sampling, we have therefore added a brief but explicit discussion of both possible volatilization losses and gas-to-filter artifact formation, and clarified how these uncertainties may affect absolute concentrations and difference-derived WSON.

Revised text in the Method on page 7, lines 186–193:

“Furthermore, integrated filter sampling may be affected by gas–particle sampling artifacts, including volatilization losses of semi-volatile inorganic nitrogen species and possible adsorption of gaseous nitrogen compounds on the filter. Previous studies suggested that adsorption of gaseous organics onto quartz filters may have only a limited effect on WSON measurement under similar sampling conditions, whereas volatilization loss during sampling may still lead to underestimation of particulate WSON (Matsumoto et al., 2014; Matsumoto and Yamato, 2016).”

3. *To show the relevance of biogenic marine WSON contribution (in abstract and main text) you should use the % contribution to total WSN, not only the contribution for each air mass.*

**Response:** Thank you for this suggestion. We have revised the relevant text throughout the manuscript.

Revised text in the Abstract on page 2, lines 40–46:

“Positive matrix factorization revealed that the contribution of sea spray aerosol (SSA)-associated WSON to total WSON increased markedly with oceanic influence,

accounting for  $3.8\% \pm 6.4\%$ ,  $14\% \pm 14\%$ , and  $34\% \pm 17\%$  under continental, mixed, and marine conditions, respectively. The corresponding contributions to WSTN were approximately  $1.6\% \pm 2.1\%$ ,  $7.3\% \pm 7.6\%$ , and  $13\% \pm 8.2\%$ , with an overall mean of  $7.8\% \pm 8.2\%$  over the sampled annual cycle.”

Revised text on page 16, lines 482–487.

“Expressed relative to total WSTN, SSA-associated WSON contributed approximately  $1.6\% \pm 2.1\%$ ,  $7.3\% \pm 7.6\%$ , and  $13\% \pm 8.2\%$  under continental-, mixed-, and marine-influenced conditions, respectively, with an overall mean contribution of  $7.8\% \pm 8.2\%$  over the sampled annual cycle, further illustrating its enhanced importance during marine influence.”

4. *The fact that WSON or WSIN are high in marine air masses does not necessarily mean that these have a marine biogenic origin. IN many studies at coastal sites using receptor modelling with PMF, there are ‘aged marine’ sources enriched in SIA and sometimes in SOA, and high loads of SIA are attributed to i) the interaction of anthropogenic  $H_2SO_4$ ,  $HNO_3$  and  $NH_4NO_3$  and  $NH_4HSO_4/(NH_4)_2SO_4$  with NaCl, interaction of anthropogenic SOA and N species may also occur; ii) Shipping emissions are anthropogenic and these may account for large emissions of SOA’s and SIA’s precursors that may be enriched in marine air masses; and iii) transport of anthropogenically polluted air masses may increase the background of SOA and SIA in marine air masses.*

**Response:** Thank you for this comment. In the original version, we mixed up the difference between the influence of the marine influence and the direct marine-biogenic attribution. We have now corrected that. The revised manuscript no longer treats elevated WSON or WSIN under marine influence as direct evidence of marine-biogenic origin; instead, we explicitly consider aged marine aerosol, shipping emissions, and polluted air masses transported over the ocean as plausible co-occurring influences.

Revised text in the Abstract on page 2, lines 50–56:

“These results provide multiple lines of evidence that SSA-associated WSON is an important contributor to coastal aerosol WSON under marine influence, with patterns consistent with marine-biogenic enhancement, although anthropogenic co-influences cannot be fully excluded.”

Added text on page 9, lines 246–250:

“It should be noted that marine-influenced air masses, as defined by low  $R_{land}$ , do not necessarily represent purely marine-biogenic conditions, because aged marine aerosol,

shipping emissions, and anthropogenically polluted air masses transported over the ocean may also contribute to aerosol composition.”

Revised text on page 13, lines 374–382:

“This pattern shows that the WSON/WSTN ratio alone may not be a reliable way to identify the source of this dataset. The elevated ratio under mixed conditions likely reflects overlapping marine and continental influences together with different responses of WSON and WSIN to transport and removal processes, rather than a unique source type. During marine-influenced periods, WSON may reflect both marine-related contributions and anthropogenic inputs associated with shipping emissions and atmospheric processing, whereas continental periods are more strongly affected by terrestrial anthropogenic emissions.”

Added text on page 17, lines 499–504:

“Although the PMF-resolved shipping factor remained low during marine-influenced periods, other anthropogenic-related factors, including secondary sulfate, VEFC and SOA factor, still showed non-negligible contributions, consistent with the view that marine-influenced air masses in this study should not be interpreted as purely marine-biogenic conditions.”

Revised text on page 20, lines 562–568:

“However, AEC may also covary with marine transport conditions, meteorology, and other seasonally structured processes, and correlation alone does not establish source dominance. Taken together, the PMF results, reduced terrestrial influence, and the positive AEC relationship are consistent with an important marine-biogenic enhancement of SSA-associated WSON during marine-influenced periods, although shipping and other anthropogenic co-influences cannot be fully excluded.”

5. *Co-linearity of PM contributing sources (shipping and biogenic precursors for example) or insolation causing high T, and high SOA and SIA production together with high emissions from photosynthesis of plankton, ... Should be considered in the discussions.*

**Response:** Thank you for this important comment. The AEC–WSON relationship should not be presented as an isolated causal effect. In a coastal atmosphere, marine influence, transport conditions, insolation, temperature, and secondary aerosol formation can co-vary. Thus, we interpret the enhanced SSA-associated WSON under

marine influence as being consistent with marine-biogenic enhancement, while explicitly acknowledging that transport conditions, atmospheric processing, and anthropogenic co-influences may also contribute.

Revised text on page 20, lines 562–568:

“However, AEC may also covary with marine transport conditions, meteorology, and other seasonally structured processes, and correlation alone does not establish source dominance. Taken together, the PMF results, reduced terrestrial influence, and the positive AEC relationship are consistent with an important marine-biogenic enhancement of SSA-associated WSON during marine-influenced periods, although shipping and other anthropogenic co-influences cannot be fully excluded.”

6. *The N isotope analyses would have provided a more solid support for the biogenic and anthropogenic origin.*

**Response:** Nitrogen isotope evidence would have made the source discussion much more robust. Unfortunately, isotopic analysis was not available for this campaign. We therefore explicitly acknowledge this as a limitation in the revised manuscript and identify isotope-based characterization as an important priority for future work.

Added text on page 22, lines 624–630:

“However, these implications should be interpreted with caution because the present dataset was obtained with seasonally uneven sampling coverage, 24 h integrated filter sampling, and without isotopic constraints that could more directly distinguish marine-biogenic, shipping-related, and continental anthropogenic sources. Future work combining more temporally uniform observations with isotopic and molecular-level characterization is needed to further strengthen source attribution.”

7. *1-5 above should be deeply discussed and results and conclusions modulated according these.*

**Response:** Thank you for these comments and suggestions. We have revised the manuscript throughout accordingly. Specifically, we have moderated the interpretation of marine-biogenic origin, clarified sampling and representativeness limitations, added SSA-associated WSON contributions relative to total WSTN, and expanded the discussion of aged marine aerosol, shipping, polluted marine background, and possible co-linearity among source and meteorological factors. The results and conclusions have therefore been revised to better reflect these uncertainties and limitations.

## Specific

### Abstract

*R35-37. This argument on supporting a ‘persistent marine source’ is not convincing me on the relevance of marine biogenic WSON (see general comments 3 and 4. If the relative proportion is the same that one in continental air masses, why the source should be different?*

**Response:** Thank you for raising this point. This statement was arguably too strong and potentially misleading. The WSON/WSTN ratio alone cannot distinguish marine from anthropogenic sources. We have therefore removed the phrase “persistent marine source” and revised the sentence accordingly.

Revised text on page 2, lines 36–40:

“Nevertheless, WSON remained a substantial fraction of water-soluble total nitrogen (WSTN) across all air-mass categories, although the WSON/WSTN ratio alone did not uniquely distinguish marine from anthropogenic influence.”

*R39-40: See comment 1 and add information to contribution of WSON of SSA-PMF to total WSN and WSON (average of the year, but taking into account comment 1).*

**Response:** Thank you for the comment. Reporting only the contribution of SSA-associated WSON to total WSON was indeed insufficient to fully evaluate its role in the overall water-soluble nitrogen budget. In the revised manuscript, we now clarify that the PMF percentages originally reported refer to the contribution of SSA-associated WSON to total WSON, and we additionally provide its contribution to total WSTN. We also avoid presenting these values as strictly climatological annual means, given the seasonally uneven sampling coverage noted above.

Revised text on page 2, lines 40–46:

“Positive matrix factorization revealed that the contribution of sea spray aerosol (SSA)-associated WSON to total WSON increased markedly with oceanic influence, accounting for  $3.8\% \pm 6.4\%$ ,  $14\% \pm 14\%$ , and  $34\% \pm 17\%$  under continental, mixed, and marine conditions, respectively. The corresponding contributions to WSTN were approximately  $1.6\% \pm 2.1\%$ ,  $7.3\% \pm 7.6\%$ , and  $13\% \pm 8.2\%$ , with an overall mean of  $7.8\% \pm 8.2\%$  over the sampled annual cycle.”

Revised text on page 16, lines 482–487:

“Expressed relative to total WSTN, SSA-associated WSON contributed approximately  $1.6\% \pm 2.1\%$ ,  $7.3\% \pm 7.6\%$ , and  $13\% \pm 8.2\%$  under continental-, mixed-, and marine-influenced conditions, respectively, with an overall mean contribution of  $7.8\% \pm 8.2\%$  over the sampled annual cycle, further illustrating its enhanced importance during marine influence.”

*R40: % contribution to what? Total N, total WSN, total WSON?*

**Response:** We now explicitly specify that these percentages refer to total WSON, and we additionally provide the contributions to total WSTN.

*Abstract general: modulate results and conclusions based on general comments 1 to 5)*

**Response:** We have revised the Abstract accordingly. In the revised version, we have moderated statements that were previously too strong, clarified that the reported PMF percentages refer to contributions to total WSON and additionally provided the corresponding contributions to total WSTN, removed wording implying a persistent or direct marine-biogenic source, and revised the concluding statement to better reflect the remaining uncertainties and possible anthropogenic co-influences. We also use the wording “over the sampled annual cycle” to avoid implying a strictly climatological annual mean.

The latest version of the abstract is as follows:

“Organic nitrogen (ON) deposition from aerosols plays a crucial role in oceanic ecosystems; however, the influence of marine biogenic activity on atmospheric ON remains poorly understood. Here, we investigate the contribution of the marine biosphere to water-soluble ON (WSON) in coastal aerosols based on particulate matter samples collected in Bangkok, Thailand, from January 2016 to January 2017. Concentrations of WSON and water-soluble inorganic nitrogen (WSIN, including  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ) were analyzed and compared across days classified by air mass origin over land as marine-, mixed-, or continental-influenced. Air masses of marine origin showed significantly lower WSON and WSIN concentrations than those from mixed and continental origins. Nevertheless, WSON remained a substantial fraction of water-soluble total nitrogen (WSTN) across all air-mass categories, although the WSON/WSTN ratio alone did not uniquely distinguish marine from anthropogenic

influence. Positive matrix factorization revealed that the contribution of sea spray aerosol (SSA)-associated WSON to total WSON increased markedly with oceanic influence, accounting for  $3.8\% \pm 6.4\%$ ,  $14\% \pm 14\%$ , and  $34\% \pm 17\%$  under continental, mixed, and marine conditions, respectively. The corresponding contributions to WSTN were approximately  $1.6\% \pm 2.1\%$ ,  $7.3\% \pm 7.6\%$ , and  $13\% \pm 8.2\%$ , with an overall mean of  $7.8\% \pm 8.2\%$  over the sampled annual cycle. Moreover, marine productivity, assessed via air mass exposure to chlorophyll-a concentrations, exhibited a strong positive correlation with SSA-associated WSON ( $r = 0.96$ ,  $p < 0.001$ ), a pattern further supported by large-scale comparison across coastal sites. These results provide multiple lines of evidence that SSA-associated WSON is an important contributor to coastal aerosol WSON under marine influence, with patterns consistent with marine-biogenic enhancement, although anthropogenic co-influences cannot be fully excluded.”

## Introduction

*R82-R87. Yes, this is the main challenge. State how do you intend to overcome limitations.*

**Response:** Thank you for this useful reminder. We have now made our strategy more explicit in the Introduction: the study does not rely on any single line of evidence, but combines PMF, back trajectories,  $R_{\text{land}}$ , and chlorophyll-a exposure to constrain source interpretation from several complementary angles. At the same time, we also state more clearly that this combination reduces, but does not eliminate, source ambiguity.

Revised text on page 4, lines 97–104:

“This discrepancy highlights the continuing challenge of distinguishing marine from anthropogenic WSON sources in coastal and adjacent marine environments. In this study, we address this issue by combining Positive Matrix Factorization (PMF) source apportionment, air-mass trajectory analysis, a trajectory-based land-retention index, and chlorophyll-a (Chl-a) exposure as complementary lines of evidence, while recognizing that these approaches reduce but do not fully eliminate source ambiguity.”

*R91 better define eutrophication as: excessive richness of nutrients in a body of water, affecting ecosystems, or something similar, not only the increase of N concentrations.*



**Response:** Thank you for this suggestion. We have revised the definition.

Revised text on page 4, lines 108–113:

“Eutrophication, defined as the excessive enrichment of aquatic systems by nutrients that alters ecosystem structure and function, may enhance primary productivity and potentially promote the emission of ON to the atmosphere (Altieri et al., 2016).”

## MATERIALS AND METHODS

*R107: Sampling 24 h. See comment 2 above discuss how the loss or gains*

**Response:** We have now clarified this point in the revised Methods. Specifically, we added a brief discussion that 24 h integrated filter sampling may be affected by gas–particle sampling artifacts, including volatilization losses of semi-volatile nitrogen species and possible uptake of gaseous species by the filter, which may influence the absolute concentrations of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and difference-calculated WSON. We also clarified that the sampling frequency was seasonally uneven, so the dataset should be interpreted as a sampled annual cycle rather than a uniformly sampled annual climatology.

Added text on page 5, lines 134–139:

“Because the sampling frequency varied among seasons and was reduced during the rainy period, this dataset does not represent a uniformly sampled annual climatology. Accordingly, the results are interpreted as observation-based estimates for the sampled annual cycle, and caution is needed when extending them to annual-scale representativeness.”

Revised text on page 7, lines 186–193:

“Furthermore, integrated filter sampling may be affected by gas–particle sampling artifacts, including volatilization losses of semi-volatile inorganic nitrogen species and possible adsorption of gaseous nitrogen compounds on the filter. Previous studies suggested that adsorption of gaseous organics onto quartz filters may have only a limited effect on WSON measurement under similar sampling conditions, whereas volatilization loss during sampling may still lead to underestimation of particulate WSON (Matsumoto et al., 2014; Matsumoto and Yamato, 2016).”

*R110-R115 Please discuss the issue of comment 1, and give the n. of samples per season.*

**Response:** We have revised the Methods accordingly. We now explicitly clarify that the sampling frequency was seasonally uneven, with reduced coverage during the rainy period, so the dataset should be interpreted as a sampled annual cycle rather than a uniformly sampled annual climatology. We have also added the number of samples collected in each season (Dry I: n = 19; Wet: n = 35; Dry II: n = 30) to make the temporal coverage more explicit.

Revised text on page 4–5, lines 128–139:

“The collection period spanned from 18 January 2016 to 28 January 2017, covering three distinct seasons: Dry I (January–March 2016, n = 19), Wet (April–June and October 2016, n = 35), and Dry II (November 2016–January 2017, n = 30). Sampling frequency averaged  $5 \pm 2$  days per month during January–February 2016 and January 2017, with intensified campaigns in March–May and late October–December. Sampling was limited between June and October due to heavy rainfall. Because the sampling frequency varied among seasons and was reduced during the rainy period, this dataset does not represent a uniformly sampled annual climatology. Accordingly, the results are interpreted as observation-based estimates for the sampled annual cycle, and caution is needed when extending them to annual-scale representativeness.”

*R140 define SOA, you will used it.*

**Response:** Corrected. “secondary organic aerosol (SOA)” is now defined at first appearance.

Revised text on page 7, lines 166.

*R151 use OC that you defined and N instead of nitrogen*

**Response:** Corrected for consistency.

Revised text on page 7, lines 178–179.

*R213 nss  $\text{SO}_4^{2-}$  (non sea salt  $\text{SO}_4^{2-}$ ) points to major marine origin? Was it ss  $\text{SO}_4^{2-}$  instead?*

**Response:** Thank you for this important correction. The original wording was indeed oversimplified. nss- $\text{SO}_4^{2-}$  is not equivalent to sea-salt sulfate, and it may include contributions from both anthropogenic sulfur and marine biogenic sulfur (Savoie et al., 2002). Our earlier result was based on the higher nss- $\text{SO}_4^{2-}$  concentrations observed

under marine-influenced conditions (Table S2). However, in the absence of more specific tracers,  $\text{nss-SO}_4^{2-}$  alone cannot be used as direct evidence of predominantly marine origin in this study. We have therefore revised the relevant passage to avoid overinterpreting  $\text{nss-SO}_4^{2-}$ , and now rely mainly on  $\text{Na}^+$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ , and  $\text{Na}^+/\Sigma\text{ions}$  as indicators of enhanced sea-salt influence, while treating  $\text{nss-SO}_4^{2-}$  and  $\text{NO}_3^-$  more cautiously as species that may also reflect secondary atmospheric processing and anthropogenic influence.

Revised text on page 9–10, lines 250–274:

“This classification is further supported by molecular marker analysis: during marine-influenced periods, the regression slope for  $\text{Na}^+$  with  $\text{Mg}^{2+}$  (0.11) closely aligned with the seawater reference ratio (0.12). Elevated levels of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ , and the  $\text{Na}^+/\Sigma\text{ions}$  ratio consistently reflected enhanced sea-salt influence during marine-influenced periods. By contrast, non-sea-salt sulfate ( $\text{nss-SO}_4^{2-}$ ) was not interpreted here as a unique indicator of marine origin, because it may include contributions from both anthropogenic sulfur and marine biogenic sulfur and may also reflect secondary atmospheric processing during transport (Savoie et al., 2002). Particulate  $\text{NO}_3^-$  was mainly interpreted as a secondary product formed from the oxidation of  $\text{NO}_x$  emitted by combustion-related sources, including traffic, shipping, industrial activities, and fossil-fuel or BB, followed by gas-to-particle partitioning or heterogeneous reactions with sea-salt particles during transport (Pryor and Sørensen, 2000). Its concentration was lowest during marine-influenced periods, lower than under mixed- and continental-influenced conditions, a pattern consistent with combustion-derived species such as non-sea-salt  $\text{K}^+$  ( $\text{nss-K}^+$ ), EC, and levoglucosan. These results indicate reduced, but not absent, terrestrial and anthropogenic influence during marine-influenced periods (Table S2).”

*R213 what about  $\text{NO}_3^-$ ?*

**Response:** Thank you for pointing this out. We have now clarified the role of  $\text{NO}_3^-$  in this section. Particulate  $\text{NO}_3^-$  was interpreted mainly as a secondary product formed from the oxidation of  $\text{NO}_x$  emitted by combustion-related sources, including traffic, shipping, industrial activities, fossil-fuel combustion, and biomass burning, followed by gas-to-particle partitioning or heterogeneous reactions with sea-salt particles during transport (Pryor and Sørensen, 2000). In our dataset,  $\text{NO}_3^-$  concentrations were lowest during marine-influenced periods and lower than those under mixed- and continental-influenced conditions, a pattern consistent with the reduced levels of combustion-derived tracers such as  $\text{nss-K}^+$ , EC, and levoglucosan. We have therefore revised the

text to make clear that  $\text{NO}_3^-$  supports reduced, but not absent, terrestrial and anthropogenic influence during marine-influenced periods.

The related discussion in this section has also been revised accordingly (9–10, lines 250–274).

*R214-216, although the long range atmospheric transport may contribute to  $\text{nssSO}_4^{2-}$  ????????  $\text{nss-SO}_4^{2-}$  is anthropogenic!!!, why biogenic sources prevailed? Rewrite*

**Response:** Thank you for your comment. Our earlier interpretation was based on the higher  $\text{nss-SO}_4^{2-}$  concentrations observed under marine-influenced conditions (Table S2). However, as clarified in the previous two comments,  $\text{nss-SO}_4^{2-}$  can include both anthropogenic and marine biogenic sulfur contributions (e.g., Savoie et al., 2002) and therefore cannot be used alone to infer that biogenic sources prevailed. We have revised this part accordingly.

The related discussion in this section has also been revised accordingly (9–10, lines 250–274).

## RESULTS

*R270-R273, is an increase from 2.7% to 3.8% in the Mediterranean compared this study is enough to underscore the relevance in this region compared to the first?*

**Response:** The difference between 2.7% and 3.8% is modest and was intended only as descriptive context rather than evidence of stronger regional relevance. We have therefore revised the sentence to a more neutral comparison without overinterpretation.

Revised text on page 11, lines 329–330:

“The mass fraction of WSTN in TSP collected in Bangkok averaged  $3.8\% \pm 1.1\%$  ( $2.2\%–7.2\%$ ), which was somewhat higher than that reported in the Eastern Mediterranean ( $\sim 2.7\%$ ; Tripathi et al., 2021) and comparable to values from Sapporo, Japan ( $3.81\% \pm 2.28\%$ ; Pavuluri et al., 2015).”

*R280-R297. Very speculative conclusions or interpretations. Please consider general comments 4 and 5 here.*

**Response:** Thank you for the comment. The original discussion in this part moved too quickly from observed contrasts to source-level interpretation. In the revised manuscript, we have retained the observed differences among air-mass regimes but moderated the associated interpretations. In particular, we no longer infer specific source origins directly from the lower or higher concentrations observed under marine influence, and we now describe wet scavenging only as one possible contributing factor rather than a direct explanation of the observed WSON patterns.

Revised text on page 12–13, lines 344–371:

“Concentrations of WSON,  $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N, TSP, OC, and EC varied considerably under different air mass regimes (Table S2). The nonparametric Mann–Whitney U test indicated that WSON and  $\text{NO}_3^-$ -N levels were significantly lower during marine-influenced periods than under mixed or continental conditions (Figure 2c,  $p < 0.001$ ). In contrast,  $\text{NH}_4^+$ -N concentrations were slightly higher during marine periods. This contrast indicates that the responses of individual nitrogen species were not uniform across air-mass regimes and should not be attributed to a single dominant source. This aligns with earlier studies reporting that aerosols in remote marine regions may still be substantially influenced by continental inputs (Jickells et al., 2013). BB tracers (e.g., levoglucosan, galactosan, mannosan) were also significantly lower during marine-influenced days (see Table S2). Furthermore, WSON correlated with BB and SOA markers and aerosol liquid water content (ALWC, Text S3) under mixed and continental conditions, whereas these associations were not evident during marine periods (Figure S7). Taken together, these patterns suggest that the lower WSON concentrations during marine-influenced periods likely reflected a combination of reduced continental and combustion-related influence, differences in transport history, and atmospheric processing. Seasonally higher rainfall may also have contributed, but because no significant direct correlation was found between WSON and precipitation (Figure S5b), precipitation alone cannot explain the observed WSON variability.”

*R298-301: Why higher under mixed air masses and very similar with marine and continental*

**Response:** We now clarify that mixed air masses likely reflect overlapping marine and continental influences, together with different responses of WSON and WSTN to transport and removal processes, so that the WSON/WSTN ratio can be elevated without pointing to a unique source type. We also state more explicitly that the WSON/WSTN ratio alone is not source-diagnostic.

Revised text on page 13, lines 372–379:

“The WSON/WSTN ratio during marine-influenced days ( $50\% \pm 17\%$ ) was similar to that under continental influence ( $48\% \pm 15\%$ ) but lower than during mixed conditions ( $60\% \pm 17\%$ ) (Figure 2d). This pattern shows that the WSON/WSTN ratio alone may not be a reliable way to identify the source of this dataset. The elevated ratio under mixed conditions likely reflects overlapping marine and continental influences together with different responses of WSON and WSIN to transport and removal processes, rather than a unique source type.”

*R300-303: You say that when coming from continental air masses the influence of anthropogenic emissions is high, but in the marine ones biogenic and shipping are the main. Shipping is anthropogenic. Furthermore, you attribute shipping emissions only to the harbour activities, and the marine transport is contributing a lot to NOx shipping emissions, not only during the periods staying in the harbours. That’s why NECA areas are related to abate NOx as O<sub>3</sub> and PM precursors.*

**Response:** Thank you for raising this point. Shipping emissions are anthropogenic and should not be grouped with marine-biogenic contributions. Moreover, shipping influence is not limited to harbor activities, because emissions from marine traffic along shipping routes can contribute substantially to NO<sub>x</sub> and related secondary aerosol formation in marine-influenced air masses. We have therefore revised the relevant text to distinguish marine-biogenic influence from shipping-related anthropogenic influence more clearly and to avoid attributing shipping effects only to harbor emissions.

Revised text on page 13, lines 379–382:

“During marine-influenced periods, WSON may reflect both marine-related contributions and anthropogenic inputs associated with shipping emissions and atmospheric processing, whereas continental periods are more strongly affected by terrestrial anthropogenic emissions.”

*R305: NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> instead of ammonium and nitrate*

**Response:** Corrected.

*R315-316: Continental region like Hawaii islands?*

**Response:** Referring to Hawaii as a continental region was inappropriate. We have revised the phrasing to distinguish receptor regions such as Hawaii from polluted continental urban regions such as Xi’an.

Revised text on page 13–14, lines 401–404:

“as well as receptor regions such as Hawaii (64%; Cornell et al., 2001) and polluted continental urban regions such as Xi’an (45%, range: 22–68%; Ho et al., 2015).”

*R318: marine background by anthropogenically polluted background*

**Response:** The original phrasing was imprecise. We have revised the text to distinguish the more marine-influenced conditions of the South China Sea from the stronger continental/anthropogenic influence over the Yellow Sea, following the comparison reported by Shi et al. (2010).

Revised text on page 14, lines 404–408:

“By comparison, the South China Sea, which is more strongly influenced by open-marine conditions, exhibited a WSON/WSTN ratio of 34%, whereas the Yellow Sea, subject to stronger continental and anthropogenic influence, showed a lower ratio of 17% (Shi et al., 2010).”

*R320 and R326 use ON acronym*

**Response:** Corrected throughout.

*R321 use WSTN acronym*

**Response:** Corrected throughout.

*R323-R324, what about the shipping emissions of in addition of the biogenic ones. Numerous ships emit a lot of NO<sub>x</sub> during their movements into the sea.*

**Response:** We now explicitly include shipping emissions in this discussion.

Revised text on page 14, lines 411–414:

“Collectively, these studies indicate that elevated WSON/WSTN ratios may arise from anthropogenic combustion sources, shipping emissions, marine-related emissions, and secondary atmospheric processing.”

*R340. shipping instead of ship, SO<sub>4</sub><sup>2-</sup> instead of sulfate*

**Response:** We have revised ship to shipping. For secondary sulfate, we retain this phrasing because it refers to the PMF factor name rather than the individual ion species. The factor was identified by high  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  loadings and is therefore interpreted as a secondary sulfate-related factor. We use  $\text{SO}_4^{2-}$  when referring to the measured ionic species.

*R342 fossil fuel combustion instead of fossil combustion*

**Response:** Corrected.

*R343,  $\text{Mg}^{2+}$  instead of Mg*

**Response:** Corrected.

*The whole page 9: There are also highly anthropogenically polluted regions by emissions from the continent and also by the shipping emissions, especially in this region with high shipping density. This can be enriched in marine air masses.*

**Response:** Based on your suggestions, we have revised the related discussion to clarify that low  $R_{\text{land}}$  indicates stronger marine influence, but does not guarantee purely marine-biogenic aerosol composition. In the revised manuscript, we explicitly acknowledge that marine-influenced air masses may still contain aged marine aerosol, shipping-related anthropogenic influence, and polluted background transported over the ocean. Accordingly, we no longer interpret elevated WSON or related ratios under marine influence as direct evidence of purely marine-biogenic origin.

Revised text on page 9, lines 246–250:

“It should be noted that marine-influenced air masses, as defined by low  $R_{\text{land}}$ , do not necessarily represent purely marine-biogenic conditions, because aged marine aerosol, shipping emissions, and anthropogenically polluted air masses transported over the ocean may also contribute to aerosol composition.”

Revised text on page 13, lines 379–382:

“During marine-influenced periods, WSON may reflect both marine-related contributions and anthropogenic inputs associated with shipping emissions and atmospheric processing, whereas continental periods are more strongly affected by terrestrial anthropogenic emissions.”



Revised text on page 16–17, lines 488–504:

“Temporal variations in source-resolved WSON concentrations are shown in Figure 3a. Among the three air mass regimes, SSA-associated WSON concentrations peaked under marine influence ( $0.19 \pm 0.12 \mu\text{gN m}^{-3}$ ), approximately 1.7 times higher than during mixed periods ( $0.11 \pm 0.12 \mu\text{gN m}^{-3}$ ) and five times higher than during continental periods ( $0.037 \pm 0.069 \mu\text{gN m}^{-3}$ ). Shipping-emission-associated WSON was also elevated during marine days ( $0.015 \pm 0.0075 \mu\text{gN m}^{-3}$ ) relative to mixed and continental periods, though its overall contribution remained low ( $\sim 3\%$ ). WSON associated with the secondary sulfate factor under marine influence ( $0.094 \pm 0.086 \mu\text{gN m}^{-3}$ ) was significantly higher than during mixed periods ( $0.022 \pm 0.023 \mu\text{gN m}^{-3}$ ) and during continental periods ( $0.046 \pm 0.056 \mu\text{gN m}^{-3}$ ), consistent with an important contribution from secondary inorganic aerosol formation. Although the PMF-resolved shipping factor remained low during marine-influenced periods, other anthropogenic-related factors, including secondary sulfate, VEFC and SOA factor, still showed non-negligible contributions, consistent with the view that marine-influenced air masses in this study should not be interpreted as purely marine-biogenic conditions.”

Revised text on page 20, lines 565–571:

“However, AEC may also covary with marine transport conditions, meteorology, and other seasonally structured processes, and correlation alone does not establish source dominance. Taken together, the PMF results, reduced terrestrial influence, and the positive AEC relationship are consistent with an important marine-biogenic enhancement of SSA-associated WSON during marine-influenced periods, although shipping and other anthropogenic co-influences cannot be fully excluded.”

*Page 9: You need to describe for the 7 factors, their chemical profile and show major and minor components for each factor. Specially for SSA, what other components it brings if you attribute all these to biogenic origin?*

**Response:** The description for the 7 factors was previously detailed in Text S1 of *Supplement*. We have expanded the PMF discussion to better describe the chemical profile of each factor and its major marker species. In particular, we clarify that the SSA factor is interpreted here as sea-salt-associated aerosol carrying WSON, rather than as purely biogenic aerosol.

Revised text on page 15, lines 431–439:

“The model resolved WSON into seven source factors: shipping emissions, secondary sulfate, dust, SOA, BB, vehicle emissions and fossil-fuel combustion

(VEFC), and SSA (Figure S8); the detailed identification of each factor is provided in Text S1. The SSA factor was characterized primarily by high loadings of  $\text{Na}^+$ ,  $\text{Cl}^-$ , and  $\text{Mg}^{2+}$ , with WSON also contributing to this factor. We therefore interpret this factor as a sea-salt-associated aerosol carrying WSON, rather than as purely biogenic aerosol.”

The description of the seven factors in the *Supplement* on page 3, lines 63–74:

“As shown in Figure S8, factor 1 was linked to high contribution of V, suggesting that it came from a shipping emission source (Celo et al., 2015; Viana et al., 2009). Factor 2 was characterized by high levels of  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$ , which was a secondary sulfate source. Factor 3 (dust) shows a high contribution of Mn (69 %), Fe,  $\text{Mg}^{2+}$ , and  $\text{Ca}^{2+}$ . Factor 4 was identified as SOA source because it was associated with high loadings of 2-MGA, 2-MGL, MBTCA, and p-phthalic acid. Factor 5 was linked to biomass burning, with high loadings of galactosan, mannosan, and levoglucosan,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and p-phthalic acid also had high loading for this source, as reported in the source profiles of biomass burning (Li et al., 2007). Factor 6 showed high loadings with Pb, Cu, and Zn and was identified as vehicle emissions and fossil fuel combustion. Factor 7 was associated with high levels of  $\text{Na}^+$ ,  $\text{Cl}^-$ , and  $\text{Mg}^{2+}$ , which was identified as sea spray aerosols via the bubble-busting processes (Facchini et al., 2008).”

*Page 383 to 398: IN which factors are  $\text{nssSO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{NO}_3$ , WSIBN will be mostly anthropogenic*

**Response:** Thank you for this helpful comment, because it exposed a gap between our PMF description and our later source discussion. We had not clearly stated where the major inorganic species sat within the factor structure. Based on the factor profiles in Figure S8,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  were mainly associated with the secondary sulfate factor, whereas  $\text{NO}_3^-$  showed a stronger contribution in the biomass-burning factor. This indicates that WSIN in our dataset was influenced predominantly by anthropogenic and secondary-processing-related sources. We have therefore revised the discussion in this section to clarify the PMF associations of the major inorganic species and to avoid overinterpreting marine-influenced conditions as direct evidence of marine-biogenic origin. The revised text now describes SSA-associated WSON enhancement more cautiously, while acknowledging the continued importance of anthropogenic and secondary-processing-related factors under different air-mass regimes.

Revised text on page 16–17, lines 488–515:

“Temporal variations in source-resolved WSON concentrations are shown in Figure 3a. Among the three air mass regimes, SSA-associated WSON concentrations peaked

under marine influence ( $0.19 \pm 0.12 \mu\text{gN m}^{-3}$ ), approximately 1.7 times higher than during mixed periods ( $0.11 \pm 0.12 \mu\text{gN m}^{-3}$ ) and five times higher than during continental periods ( $0.037 \pm 0.069 \mu\text{gN m}^{-3}$ ). Shipping-emission-associated WSON was also elevated during marine days ( $0.015 \pm 0.0075 \mu\text{gN m}^{-3}$ ) relative to mixed and continental periods, though its overall contribution remained low ( $\sim 3\%$ ). WSON associated with the secondary sulfate factor under marine influence ( $0.094 \pm 0.086 \mu\text{gN m}^{-3}$ ) was significantly higher than during mixed periods ( $0.022 \pm 0.023 \mu\text{gN m}^{-3}$ ) and during continental periods ( $0.046 \pm 0.056 \mu\text{gN m}^{-3}$ ), consistent with an important contribution from secondary inorganic aerosol formation. Although the PMF-resolved shipping factor remained low during marine-influenced periods, other anthropogenic-related factors, including secondary sulfate, VEFC and SOA factor, still showed non-negligible contributions, consistent with the view that marine-influenced air masses in this study should not be interpreted as purely marine-biogenic conditions. These results also indicate that marine air mass transport plays an important role in the enhancement of SSA-associated WSON, further supported by a strong negative correlation between SSA-associated WSON and  $R_{\text{land}}$  (Figure 3c,  $r = -0.59$ ,  $p < 0.001$ ). In contrast, SOA-, BB-, and VEFC-associated WSON increased significantly under mixed and continental conditions. Although SOA is generally considered more susceptible to wet removal than primary aerosol (Sun et al., 2011; Zhao et al., 2026), no significant correlation was observed between precipitation and PMF-resolved source concentrations in this dataset. This suggests that wet scavenging alone did not dominate the observed source-resolved WSON variability.”

*R359 and R410 use ON acronym.*

**Response:** Thank you for this suggestion. Corrected in R359. The section in R410 has been revised as follows: “SSA is dominated by inorganic sea salt but can also contain an important organic fraction derived from ocean-surface materials.”

*R410 SSA is a complex mixture of organic but mainly inorganic, isnt'it?*

**Response:** Thank you for this correction. We have revised the SSA description to avoid implying that SSA is predominantly organic.

Revised text on page 19, lines 528–530.

“SSA is dominated by inorganic sea salt but can also comprise an important organic fraction derived from ocean-surface materials.”

*R421 aerosol carbon by aerosol's carbon*

**Response:** Thank you for pointing this out. This sentence has been removed during revision, as we shortened this background discussion to improve the focus of this section.

## CONCLUSIONS

*Consider general comments above to draw the conclusions. Define again acronyms in this section, and when defined use only acronym.*

**Response:** Thank you for this important suggestion. We have rewritten this section accordingly. The revised version no longer claims source dominance, presents the AEC relationship more cautiously, and states the main limitations of the study in plain terms—sampling representativeness, filter-based uncertainty, and the lack of isotope constraints. We have also reintroduced the key acronyms in this section and then used them consistently.

Revised text on page 22–23, lines 603–644:

“The equation derived in this study ( $\text{WSON} [\text{nmol m}^{-3}] = 276 \times \text{AEC} [\text{mg m}^{-3}] - 2.74$ ) provides an empirical basis for examining the linkage between coastal aerosol water-soluble organic nitrogen (WSON) and oceanic biological conditions along air-mass transport pathways. Our results indicate that sea spray aerosol (SSA)-associated WSON is an important contributor to coastal aerosol WSON under marine-influenced conditions, and its covariation with trajectory-based air-mass exposure to chlorophyll-a (Chl-a) (AEC) is consistent with marine-biogenic enhancement. However, these implications should be interpreted with caution because the present dataset was obtained with seasonally uneven sampling coverage, 24 h integrated filter sampling, and without isotopic constraints that could more directly distinguish marine-biogenic, shipping-related, and continental anthropogenic sources. Future work combining more temporally uniform observations with isotopic and molecular-level characterization is needed to further strengthen source attribution. We therefore suggest that integrating satellite-derived Chl-a data into air mass trajectory analyses may help improve future assessments of marine-related WSON variability.”

## FORMAT IN ALL TEXT

*In results: Present always mean $\pm$  std (min-max). In R69 you do it like this in R275-6 you give min-max (mean $\pm$  std)*

**Response:** Thank you for this careful suggestion. We have revised the manuscript throughout to improve consistency in statistical formatting. Mean  $\pm$  standard deviation and min–max formats are now harmonized. The final presentation is: mean $\pm$  std (min–max).

*Define all acronyms, N, S, ON, WSTN, WSON, IN, WSIN,.... And once defined, use always the acronym, now these are defined in most cases (not N, nor S, ..... ) and these acronyms and complete spelling is used alternatively along the paper. Ex nitrogen (N) is not used as N but later you use ON as acronym defined in R51 but fully spelled in R55, R73, re-defined in R89,.....*

**Response:** We have revised the manuscript throughout to improve consistency in acronym use. Acronyms are defined at first appearance and used consistently thereafter.

## References:

- Pryor, S. C., and L. L. Sørensen (2000), Nitric Acid–Sea Salt Reactions: Implications for Nitrogen Deposition to Water Surfaces, *J. Appl. Meteorol.*, 39(5), 725-731. <https://doi.org/10.1175/1520-0450-39.5.725>.
- Savoie, D. L., R. Arimoto, W. C. Keene, J. M. Prospero, R. A. Duce, and J. N. Galloway (2002), Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean, *J. Geophys. Res.-Atmos.*, 107(D18), AAC 3-1-AAC 3-21. <https://doi.org/10.1029/2001JD000970>.
- Shi, J., H. Gao, J. Qi, J. Zhang, and X. Yao (2010), Sources, compositions, and distributions of water-soluble organic nitrogen in aerosols over the China Sea, *J. Geophys. Res.-Atmos.*, 115(D17). <https://doi.org/10.1029/2009JD013238>.