

1       **Marine-Derived Water-Soluble Organic Nitrogen in**  
2       **Coastal Air: Influence of Ocean Productivity on**  
3       **Atmospheric Nitrogen Cycling**

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## Abstract

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 ~~organic-nitrogen~~ 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, ~~the relative proportion of~~ WSON ~~remained a substantial fraction of~~ in water-soluble total nitrogen (WSTN) across all air-mass categories, although the WSON/WSTN ratio alone did not uniquely distinguish marine from anthropogenic influence. ~~remained consistent, implying a persistent marine source.~~ Positive matrix factorization revealed that the contribution of sea spray aerosol (SSA)-~~associated~~ derived 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~~ chlorophyll-a concentrations (~~AEC~~), exhibited a strong positive correlation with ~~SSA-derived~~ SSA-associated WSON ( $r = 0.96$ ,  $p < 0.001$ ), a ~~pattern finding~~ further supported by large-scale comparison across coastal sites ~~reanalysis~~. These results provide multiple lines of direct evidence that SSA-associated WSON ~~marine organic aerosols is an important contributor to coastal aerosol~~ represent a major source of WSON under marine influence in coastal regions globally, with patterns consistent with marine-biogenic enhancement, although anthropogenic co-influences cannot be fully excluded ~~with important implications for atmospheric nitrogen cycling and climate feedback processes.~~

## 1. Introduction

Organic nitrogen (ON), which includes compounds such as amino acids, urea, organic nitrates, nitroaromatics, and humic-like substances, plays an important role in atmospheric processes including air quality, cloud formation, and the nitrogen cycle (Cape *et al.*, 2011). On a global scale, water-soluble ~~organic nitrogen~~ WSON has been estimated to contribute 10%–40% of total airborne ON (Cape *et al.*, 2011; Liu *et al.*, 2021; Matsumoto *et al.*, 2019a; Wang *et al.*, 2018), influencing aerosol properties such as solubility, acidity, and hygroscopicity. Furthermore, certain nitrogen-containing organic compounds, including nitroaromatics, have been recognized as important chromophores in brown carbon (He *et al.*, 2022; Liu *et al.*, 2023), thereby influencing radiative forcing. In addition, atmospheric deposition of particulate WSON is increasingly regarded as a significant source of nitrogen input to marine ecosystems (Buchanan *et al.*, 2021; Li *et al.*, 2023).

WSON originates from both direct emissions—including anthropogenic and biogenic sources—and secondary formation through atmospheric reactions (Xu *et al.*, 2020; Yu *et al.*, 2017). These complex sources and atmospheric processes contribute to substantial spatial and temporal variability in WSON deposition (Kanakidou *et al.*, 2012; Li *et al.*, 2023; Yu *et al.*, 2020). Previous studies have identified marine emissions as a notable source of atmospheric ON (Facchini *et al.*, 2008; O'Dowd *et al.*, 2004). Globally, the estimated annual primary emission of soluble ON from the ocean is 2.1 Tg N yr<sup>-1</sup>, comparable in magnitude to anthropogenic emissions from fossil fuel combustion and biomass burning (BB) (Ito *et al.*, 2014; Kanakidou *et al.*, 2012). ~~In some remote marine regions, isotopic evidence suggests~~ Consequently, ~~organic nitrogen that aerosol ON in marine aerosols can be strongly influenced by is considered to be primarily derived from marine~~ biological production rather than terrestrial pollution (Altieri *et al.*, 2016).

Recent research has underscored the complexity and variability of WSON in sea spray aerosol (SSA) (Altieri *et al.*, 2012; Li *et al.*, 2019). For instance, primary emissions of ~~sea-spray emissions sea-salt particles~~ have been recognized as a major source of WSON over the remote Indian sector of the Southern Ocean (Matsumoto *et al.*, 2022). These findings highlight the importance of incorporating marine ON emissions in assessments of net atmospheric WSON deposition, particularly in the open ocean (Luo *et al.*, 2018).

However, several field studies in regions strongly influenced by marine air masses have reported only minor contributions from marine-derived WSON—typically below 5% (Leung *et al.*, 2024; Tsagkaraki *et al.*, 2021). This discrepancy highlights the continuing challenge of distinguishing marine from anthropogenic WSON sources~~indicates that distinguishing anthropogenic from marine sources remains a challenge, even~~ 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.

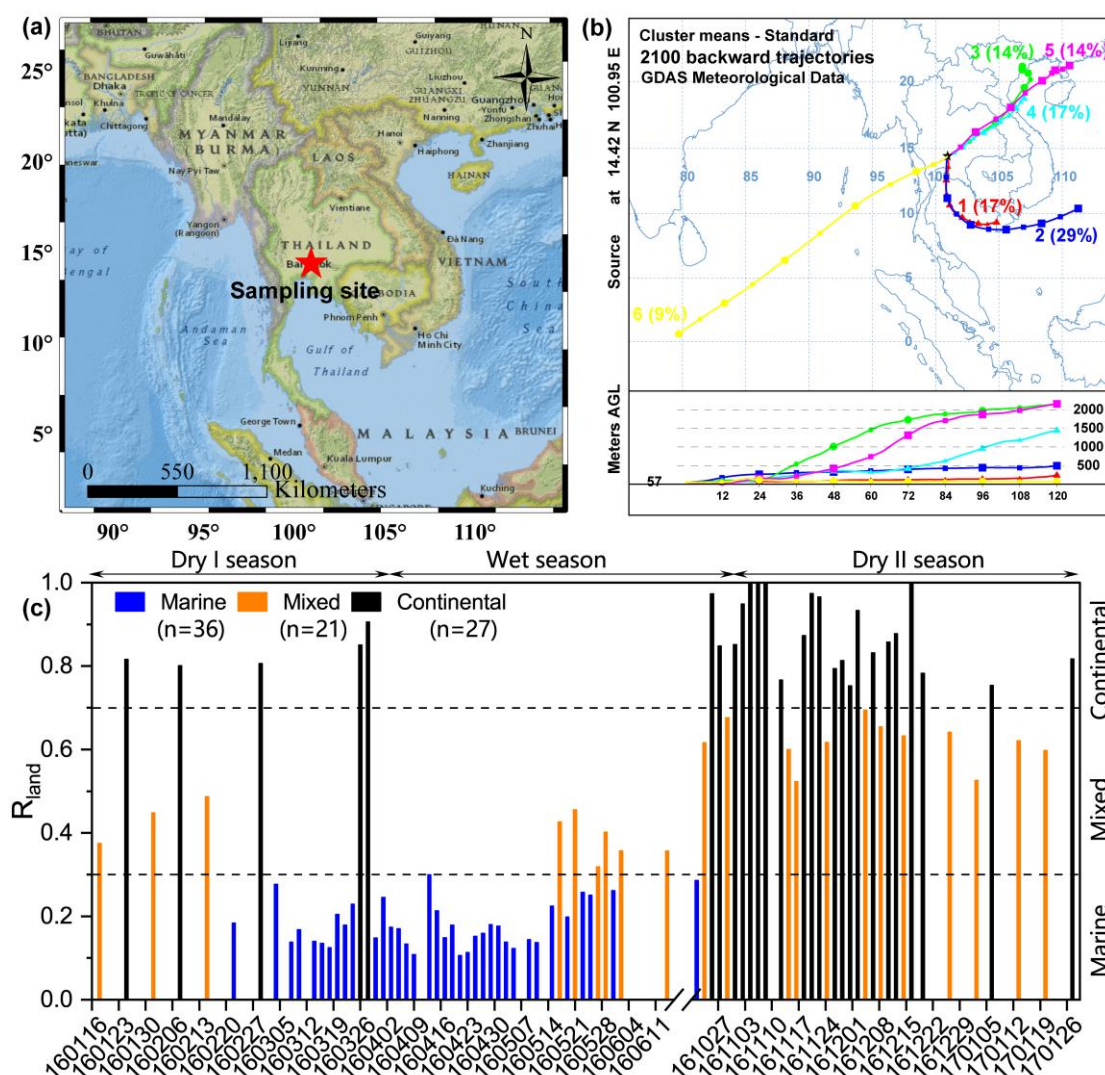
The Indochinese Peninsula (ICP), characterized by high population density and substantial ~~organic-nitrogen (ON)~~ deposition (Ito *et al.*, 2014; Kanakidou *et al.*, 2012; Li *et al.*, 2023), provides a suitable context for assessing the influence of marine aerosols on atmospheric WSON. Eutrophication, defined as the excessive enrichment of aquatic systems by nutrients that alters ecosystem structure and function,~~—the gradual enrichment of aquatic ecosystems with nutrients such as nitrogen—~~may enhance primary productivity and ~~potentially can~~ promote the ~~subsequent~~ emission of ON to the atmosphere (Altieri *et al.*, 2016). Here, we selected Bangkok, the capital of Thailand, which is situated in the central plain of the country and adjacent to the Gulf of Thailand. The region experiences prevailing marine winds from January to October, offering a favorable setting for studying marine aerosol contributions to WSON. Our study aims to: (1) quantify WSON abundances at a coastal site in the ICP; (2) assess marine influences on WSON distributions; and (3) elucidate the mechanisms governing oceanic contributions to WSON.

## 2. Material and Methods

### 2.1. Sampling Campaign

A total of 84 total suspended particulate (TSP) samples were collected from the rooftop (57 m above ground level) of the Faculty of Environment at Kasetsart University (100°57' E and 13°85' N; Figure 1a) in Bangkok, Thailand—a site previously characterized in air quality studies (Tang *et al.*, 2021; Wang *et al.*, 2020). Sampling was conducted over 24-hour periods using a high-volume air sampler (flow rate: 0.3 m<sup>3</sup> min<sup>-1</sup>) equipped with pre-combusted quartz-fiber filters (450 °C for 6 h). 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. Precipitation and solar radiation data were obtained from historical reanalysis products provided by the European Centre for Medium-Range Weather Forecasts (ECMWF). All samples and field blanks were stored in the dark at  $-20\text{ }^{\circ}\text{C}$  until analysis. This storage procedure helps minimize post-collection changes, but it does not eliminate artifacts generated during sampling itself. A summary of TSP mass concentrations, chemical components, and meteorological conditions is provided in Table S1 (*Supplement*).



**Figure 1.** (a) Sampling site location in Bangkok, Thailand. (b) Classified air mass trajectories (detailed in Figure S1–S2). (c) Distribution of ~~the air mass retention ratio over land~~ ( $R_{land}$ ), with samples categorized as marine-influenced ( $R_{land} < 0.3$ ), mixed-influenced ( $0.3 \leq R_{land} \leq 0.7$ ), or continental-influenced ( $R_{land} > 0.7$ ) based on the  $R_{land}$  values. The map in panel (a) was created using ArcGIS software with the base layer from the ESRI National Geographic World Map.

## 2.2. Chemical Analysis

Organic carbon (OC) and elemental carbon (EC) mass concentrations were determined using an OC/EC analyzer following the NIOSH\_870 thermal-optical protocol. Inorganic ions ( $Cl^-$ ,  $NO_3^-$ ,  $SO_4^{2-}$ ,  $Na^+$ ,  $K^+$ ,  $NH_4^+$ ,  $Mg^{2+}$ , and  $Ca^{2+}$ ) were quantified by ion chromatography (761 Compact IC, Metrohm, Switzerland), and trace elements were analyzed via inductively coupled plasma–mass spectrometry (ICP–MS; ELAN DRC II, PerkinElmer Ltd., Hong Kong). Analytical errors were 5.5% for OC, and 3.9% for EC, below 5.0% for



trace elements, and under 1.0% for ~~water-water~~-soluble ions, based on prior validation (Wang *et al.*, 2020).

Polar molecular tracers—including ~~biomass—burning~~BB markers (levoglucosan, mannosan, galactosan) and biogenic/anthropogenic secondary organic aerosol (~~SOA~~) tracers such as 2-methylglyceric acid (2-MGA), 2-methylthreitol and 2-methylerythritol (2-MGL), 3-methyl-1,2,3-butanetricarboxylic acid (MBTCA), and o/p-phthalic acid—were analyzed by gas chromatography–mass spectrometry (GC–MS) following derivatization as previously reported (Geng *et al.*, 2020; Li *et al.*, 2013). The mean recovery of <sup>13</sup>C-labeled levoglucosan was 87% ± 10%. Non-polar tracers of coal and fossil fuel combustion (hopanes and steranes) were also analyzed, with perdeuterated tetracosane yielding a recovery of 114% ± 11% (Wang *et al.*, 2020).

Water-soluble OC (WSOC) and water-soluble total nitrogen (WSTN) were extracted by ultrasonication for 30 minutes using ultrapure water (resistivity > 18.2  $\text{M}\Omega\cdot\text{cm}^{-1}$ ), followed by filtration through 0.22  $\mu\text{m}$  PTFE membranes. Concentrations were measured with a ~~total-organic-carbon~~TOC/~~total-nitrogen~~TN analyzer (model TOC-Vcsh, Shimadzu). WSON was calculated as the difference between WSTN and water-soluble inorganic nitrogen (WSIN), where WSIN comprises  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , and  $\text{NO}_2^-\text{-N}$ :  $[\text{WSON}] = [\text{WSTN}] - [\text{WSIN}]$ . Nitrite concentrations were consistently below the detection limit of ion chromatography and were excluded from further analysis. It should be noted that some dissolved ~~organic-nitrogen~~ON species may not be fully converted to nitrogen monoxide in the TOC/TN analyzer, potentially leading to underestimation of WSON (Miyazaki *et al.*, 2011). 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).

The relative standard deviation (RSD) for WSTN analysis ~~were~~was 3.6% (method) and 0.77% (instrument). Method detection limits were 0.09  $\mu\text{g m}^{-3}$  for WSON, 0.03  $\mu\text{g m}^{-3}$  for  $\text{NO}_3^-$ , and 0.02  $\mu\text{g m}^{-3}$  for  $\text{NH}_4^+$ . Field blank levels were

0.067  $\mu\text{gN m}^{-3}$  (WSN), 0.043  $\mu\text{gN m}^{-3}$  ( $\text{NH}_4^+\text{-N}$ ), and 0.07  $\mu\text{gN m}^{-3}$  ( $\text{NO}_3^-\text{-N}$ ), corresponding to average blank-to-sample ratios of 7.1%, 4.3%, and 12%, respectively, consistent with previous reports (Matsumoto *et al.*, 2019a). All reported WSN and WSTN concentrations were blank-corrected and should be interpreted as operationally defined particulate water-soluble N concentrations under the applied sampling protocol.

## 2.3 Source Apportionment

The U.S. Environmental Protection Agency's ~~Positive—Matrix Factorization~~PMF model (PMF 5.0) was employed to perform factor analysis on environmental data with non-negativity constraints and to estimate associated uncertainties (Norris *et al.*, 2014). PMF has been widely applied as a robust tool for aerosol source apportionment. In PMF 5.0, species are evaluated based on the signal-to-noise (S/N) ratio and can be classified as “strong,” “weak,” or “bad.” Weak species are retained but assigned a tripled uncertainty, whereas bad species are excluded from the modeling. In this study, WSN was included as a total variable to resolve its sources. Following the base run, rotational stability ( $F_{\text{peak}}$ ) tests were conducted, and model robustness was evaluated using the base model displacement (DISP), bootstrap (BS), and bootstrap displacement (BS-DISP) methods. A detailed description of PMF procedures is provided in Text S1 in *Supplement*.

## 2.4. Air Mass Back Trajectories and Trajectory-Based ~~chlorophyll a~~Chl-a Exposure.

To identify potential source regions, we calculated 120-hour back trajectories using the Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) model (<http://www.arl.noaa.gov/HYSPLIT.php>), driven by the Global Data Assimilation System (GDAS) meteorological dataset at  $1^\circ \times 1^\circ$  resolution (<http://ready.arl.noaa.gov/archives.php>). Trajectories were generated at 1-hour intervals and subsequently classified through cluster analysis (Figure 1b and Figures S1–S2). Based on origin and transport pathways, air masses arriving in Bangkok were grouped into six distinct clusters. During the Dry I season, air masses originated predominantly over the Gulf of Thailand-Bay/South China Sea (clusters 1–2), with a minor contribution from the Indochina Peninsula (cluster 3). In the Wet season, trajectories were primarily transported via the South China Sea/Gulf of Thailand-Bay and the Arabian Sea (clusters 1, 2, 6),



whereas Dry II season air masses mainly originated over mainland China and crossed the Indochina Peninsula (clusters 3–5).

Furthermore, the air mass retention ratio over land ( $R_{land}$ ), defined as the weighted ratio of transport time over land to the total transport duration, was calculated according to the method of Zhou *et al.* (2021, 2023) using Equation 1. This parameter provides a quantitative measure of terrestrial influence at the receptor site. A schematic illustration is presented in Figure S3.

$$R_{land} = \frac{\sum_{i=1}^{N_{land}} e^{-t_i/120}}{\sum_{i=1}^{N_{total}} e^{-t_i/120}} \quad (1)$$

Here,  $N_{total}$  denotes the total number of trajectory endpoints and  $N_{land}$  the number over land. The backward tracking time  $t_i$  (in hours) and the weighting factor  $e^{-t_i/120}$  account for the diminishing influence of distant regions due to air mass diffusion and particle deposition during transport. As a result, regions associated with longer backward tracking times exert a weaker influence on the receptor site compared to nearby areas. Based on the  $R_{land}$  values (Figure 1c), samples were categorized as marine-influenced ( $R_{land} < 0.3$ ), mixed-influenced ( $0.3 \leq R_{land} \leq 0.7$ ), or continental-influenced ( $R_{land} > 0.7$ ). 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. This classification is further supported by molecular marker analysis: during marine-influenced periods, the regression slope for  $Na^+$  ~~with versus~~  $Mg^{2+}$  (0.11) closely aligned with the seawater reference ratio (0.12). Elevated levels of  $Cl^-$ ,  $Na^+$ ,  ~~$Mg^{2+}$ , non-sea-salt sulfate ( $nss-SO_4^{2-}$ ), and the~~  $Na^+/\Sigma ions$  ratio consistently reflected enhanced sea-salt influence during marine-influenced periods. dominant contributions from sea spray emissions. By contrast, non-sea-salt sulfate ( $nss-SO_4^{2-}$ ) 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  $NO_3^-$  was mainly interpreted as a secondary product formed from the oxidation of  $NO_x$  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). Although long-range anthropogenic transport may contribute to nss-SO<sub>4</sub><sup>2-</sup>, marine biogenic sources prevailed, with concentrations significantly higher during marine-influenced periods than during mixed or continental periods. Its concentration was lowest during marine-influenced periods, lower than under mixed- and continental-influenced conditions, a pattern consistent with ~~contrast~~, combustion-derived species such as non-sea-salt K<sup>+</sup> (nss-K<sup>+</sup>), EC, and levoglucosan ~~showed markedly higher concentrations during continental and mixed-influenced periods~~. These results indicate reduced, but not absent, ~~terrestrial and anthropogenic influence than~~ during marine-influenced periods (Table S2).

Air mass exposure to ~~chl~~ Chl-a (AEC), defined as the mean sea surface ~~Chl-a~~ Chl-a concentration along air mass trajectories, was used as a proxy for marine biogenic emissions at the receptor site (Park *et al.*, 2018; Zhou *et al.*, 2023). A statistically significant positive correlation was observed when air masses traveled within the marine boundary layer. However, due to the relatively low correlation between AEC and methanesulfonate, the formulation was adjusted based on the approach of Zhou *et al.* (2021, 2023), as follows:

$$AEC = \frac{\sum_{i=1}^{N_{total}} Chl-a_i \cdot e^{-t_i/120} \cdot e^{-h_i/500}}{n} \quad (2)$$

Here,  $N_{total}$  denotes the total number of hourly endpoints (120, including the receptor point) along the trajectory. The variable  $Chl-a_i$  represents the mean ~~Chl-a~~ Chl-a concentration—derived from MODIS-Aqua monthly composites at a 4 km resolution—within a 20 km radius of the  $i_{th}$  trajectory endpoint. Endpoints over land were assigned  $Chl-a_i = 0$ . The weighting factor  $e^{-h_i/500}$  accounts for the influence of altitude  $h_i$ , reflecting the reduced contribution from higher altitudes due to ~~Chl-a~~ Chl-a dilution and particle deposition during transport. The denominator  $n$  corresponds to the number of trajectory endpoints with valid ~~Chl-a~~ Chl-a data, including zero values over land.

## 2.5. Potential Source Contribution Function (PSCF) Model

The PSCF model was employed to identify source regions by discretizing the study domain into an  $i \times j$  grid. The PSCF values, ranging from 0 to 1,

represent the conditional probability that an air parcel passing through a grid cell contributes to high concentrations at the receptor site; elevated values denote a higher probability of source contribution. In this study, PSCF analysis was applied to identify potential geographic source regions of both total WSON and PMF-resolved source categories of WSON in aerosols collected in Bangkok. A detailed description of the PSCF methodology is provided in Text S2 and in previous publications (Geng *et al.*, 2020; Tang *et al.*, 2024).

### 3. Results and Discussion

#### 3.1. Temporal Variations of WSON

Figure S4 and Table S1 present the temporal variations and statistical summaries of meteorological parameters and chemical compositions in TSP throughout the sampling campaign. In Bangkok, Thailand, the mass concentrations of TSP, OC, and EC ~~varied from~~ were  $55 \pm 30 \mu\text{g m}^{-3}$  (17–161  $\mu\text{g m}^{-3}$ ) (mean:  $55 \pm 30 \mu\text{g m}^{-3}$ ),  $12 \pm 6.3 \mu\text{g m}^{-3}$  (3.7–38  $\mu\text{g m}^{-3}$ ) (mean:  $12 \pm 6.3 \mu\text{g m}^{-3}$ ), and  $1.4 \pm 0.43 \mu\text{g m}^{-3}$  (0.16–2.8  $\mu\text{g m}^{-3}$ ) (mean:  $1.4 \pm 0.43 \mu\text{g m}^{-3}$ ), respectively. The TSP levels in this region were substantially lower than those reported in other areas, such as the Eastern Mediterranean ( $219.90 \pm 104.85 \mu\text{g m}^{-3}$ ; Tripathee *et al.*, 2021), Jiaozhou Bay ( $133.74 \pm 80.35 \mu\text{g m}^{-3}$ ; Xing *et al.*, 2018), and Xi'an during the dust episodes ( $2109 \pm 1360 \mu\text{g m}^{-3}$ ; Wang *et al.*, 2014). Pronounced seasonal variations were observed: TSP levels decreased by 56% from the Dry I season to the Wet season, but increased by 52% during the Dry II season. In Bangkok, Rainfall during the Wet season (April to October) accounted for 92% of the annual precipitation total. To examine wet scavenging effects, we evaluated the relationship between TSP concentrations and precipitation (Figure S5a). A significant negative correlation was identified ( $R^2 = 0.22$ ,  $p < 0.001$ ), consistent with a contribution from wet scavenging, although the relatively low explanatory power indicates that emissions, transport, and precipitation history along the air-mass pathway also substantially influenced TSP variability indicating efficient aerosol removal by rainfall. The relatively low  $R^2$ -value, however, suggests additional influences from emission sources and precipitation occurring during long-range transport.

The mass fraction of WSTN in TSP collected in Bangkok averaged  $3.8\% \pm 1.1\%$  (~~range~~  $2.2\%–7.2\%$ ), which was somewhat higher than that reported in the Eastern Mediterranean ( $\sim 2.7\%$ ; Tripathee *et al.*, 2021) and comparable to

values from Sapporo, Japan ( $3.84\% \pm 2.28\%$ ; Pavuluri *et al.*, 2015), underscoring the relevance of water-soluble nitrogen in this region. As shown in Figure 2a and Table S2, the concentrations of the individual WSTN components, including WSON,  $\text{NO}_3^-$ -N, and  $\text{NH}_4^+$ -N, ranged from  $0.95 \pm 0.40 \mu\text{gN m}^{-3}$  to  $2.3 \mu\text{gN m}^{-3}$  ( $0.27$ – $2.3 \mu\text{gN m}^{-3}$ ),  $0.60 \pm 0.52 \mu\text{gN m}^{-3}$  below detection limitation (BDL) to  $2.7 \mu\text{gN m}^{-3}$  (below detection limitation (BDL)– $2.7 \mu\text{gN m}^{-3}$ ), and  $0.47 \pm 0.44 \mu\text{gN m}^{-3}$  BDL– $2.2 \mu\text{gN m}^{-3}$  (BDL– $2.2 \mu\text{gN m}^{-3}$ ), respectively. WSON correlated positively with both TSP ( $r = 0.65$ ,  $p < 0.01$ ) and WSIN ( $r = 0.51$ ,  $p < 0.01$ ) (Figure S6), indicating that WSON variability was linked to overall aerosol loading and co-varied with inorganic N across the dataset suggesting possible common sources or formation pathways (Figure S6).

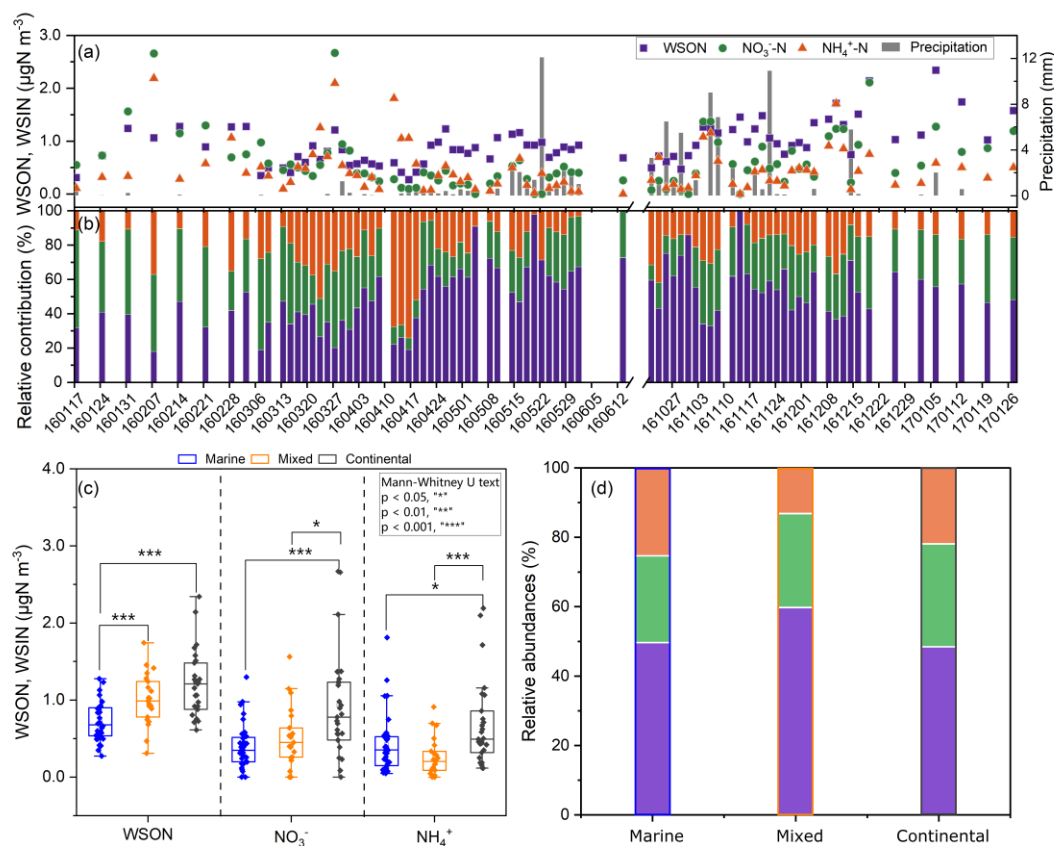
Concentrations of WSON,  $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N, TSP, OC, and EC varied considerably under different air mass regimes (Table S2). The nonparametric Mann–Whitney U test indicated that WSON and  $\text{NO}_3^-$ -N levels were significantly lower during marine-influenced periods than under mixed or continental conditions (Figure 2c,  $p < 0.001$ ). In contrast,  $\text{NH}_4^+$ -N concentrations were slightly elevated higher during marine periods. This contrast indicates that the responses of individual N species were not uniform across air-mass regimes and should not be attributed to a single dominant source consistent with the ocean potentially acting as a source of atmospheric ammonia (Altieri *et al.*, 2014). This pattern aligns with earlier studies reporting that aerosols higher WSON in aerosols of remote marine regions may still be substantially influenced by under continental inputs influence than in those of purely oceanic origin, indicating a stronger terrestrial contribution (Jickells *et al.*, 2013). Biomass burning BB tracers (e.g., levoglucosan, galactosan, mannosan) were also significantly lower during marine-influenced days (see Table S2). Furthermore, WSON correlated with biomass burning markers BB and SOA markers and aerosol liquid water content (ALWC, Text S3) under mixed and continental conditions, whereas these associations absent were not evident during marine periods (Figure S7). Wet scavenging may partly explain the lower WSON levels in marine periods, which largely coincided with the rainy season (Figure 2a), though no direct correlation was found between WSON and precipitation. 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.

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. 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. Continental WSON/WSTN ratios likely reflect anthropogenic emissions, whereas marine periods may be influenced by marine biogenic emissions or shipping activity from Bangkok Port. Mixed conditions likely represent combined source influences. Precipitation may alter the WSON/WSTN ratio through differential scavenging: WSIN species (e.g.,  $\text{NO}_3^-$ -nitrate,  $\text{NH}_4^+$ -ammonium) are efficiently removed by rainfall (Matsumoto *et al.*, 2019b; Nehir and Koçak, 2018), as WSIN shown-shown by their stronger correlation with precipitation (Figure S5c,d), whereas the lack of such a correlation for WSON suggests distinct scavenging behavior. By contrast, the absence of a significant WSON-precipitation correlation indicates that WSON variability in this dataset was less directly coupled to precipitation, rather than supporting a distinct scavenging mechanism.

Annually, WSON accounted for  $52\% \pm 17\%$  of the WSTN (Figure 2b)—substantially higher than values reported from a forest site ( $20\% \pm 11\%$ ; Miyazaki *et al.*, 2014), an offshore island (27%; Tian *et al.*, 2023), Sapporo ( $9.2\% \pm 7.3\%$ ; Pavuluri *et al.*, 2015), and coastal Qingdao ( $\sim 20\%$ ; Shi *et al.*, 2010). Elevated WSON/WSTN ratios have been documented in source emissions such as biomass-burningBB ( $80\% \pm 6.3\%$ ), vehicle exhaust ( $67\% \pm 16\%$ ), and shipping emissions ( $54\% \pm 31\%$ ) (Yu *et al.*, 2017), as well as receptor regions such as in polluted continental regions like Hawaii (64%; Cornell *et al.*, 2001)

and polluted continental urban regions such as Xi'an (45%, range: 22%–68%; Ho *et al.*, 2015). By comparison, the South China Sea, which is more strongly influenced by open-marine —characterized by marine background—conditions, —exhibited a WSON/WSTN ratio of 34%, whereas higher than that of the Yellow Sea, subject to stronger continental and anthropogenic influence, showed a lower ratio of 17% —(17%; Shi *et al.*, 2010). During phytoplankton blooms, organic-nitrogen WSON can dominate aerosol composition, contributing up to 84% of total-dissolved-nitrogen WSTN (Violaki *et al.*, 2015) and 63% of submicrometer aerosol mass (O'Dowd *et al.*, 2004). Collectively, these studies indicate that elevated WSON/WSTN ratios may arise from either anthropogenic combustion influence sources, shipping emissions, or marine-biogenic activity-related emissions, and secondary atmospheric processing. Therefore, the WSON/WSTN ratio alone is insufficient to discriminate marine-biogenic, shipping-related, and continental anthropogenic sources of WSON. More quantitative approaches are needed to apportion the origins of aerosol organic nitrogen WSON.



**Figure 2.** Temporal variations and source influences on nitrogen-N species in Bangkok



aerosols. (a) Time-series of concentrations of WSON and WSIN ( $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ ), overlaid with daily rainfall from ECMWF reanalysis data. (b) Relative contributions of WSON and WSIN to ~~total water-soluble-nitrogen~~WSTN across the study period. (c) Concentration distributions and (d) relative abundances of WSON and WSIN during marine-, mixed-, and continental-influenced periods.

### 3.2. ~~Sea Spray Aerosols~~SSA as a Major WSON Source in Marine-influenced Days

To elucidate the contributions of marine and anthropogenic sources to WSON in this coastal urban environment, we applied the PMF 5.0 model to 84 aerosol samples characterized by 26 chemical species. The model resolved WSON into seven source factors: ship~~ping~~ emissions, secondary sulfate, dust, ~~secondary organic aerosol (SOA)~~, ~~biomass burning (BB)~~, vehicle emissions and ~~fossil-fossil-fuel~~ combustion (VEFC), and ~~sea spray aerosol (SSA)~~ (Figure S8); ~~the detailed identification of each factor is provided in Text S1. Here, the~~ SSA factor was ~~characterized identified 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. because we note that SSA-derived WSON was significantly associated with marine biological activity not merely sea salt particles as elaborated below.~~ Furthermore, PSCF mapping of ~~SSA-derived-associated~~ WSON ~~pointed mainly to~~was ~~mainly associated with~~ the Gulf of Thailand and, to a lesser extent, the Bay of Bengal ~~(see Figure S10), supporting the importance of marine-related source regions for this factor. reinforcing the role of sea spray as a relevant WSON source.~~

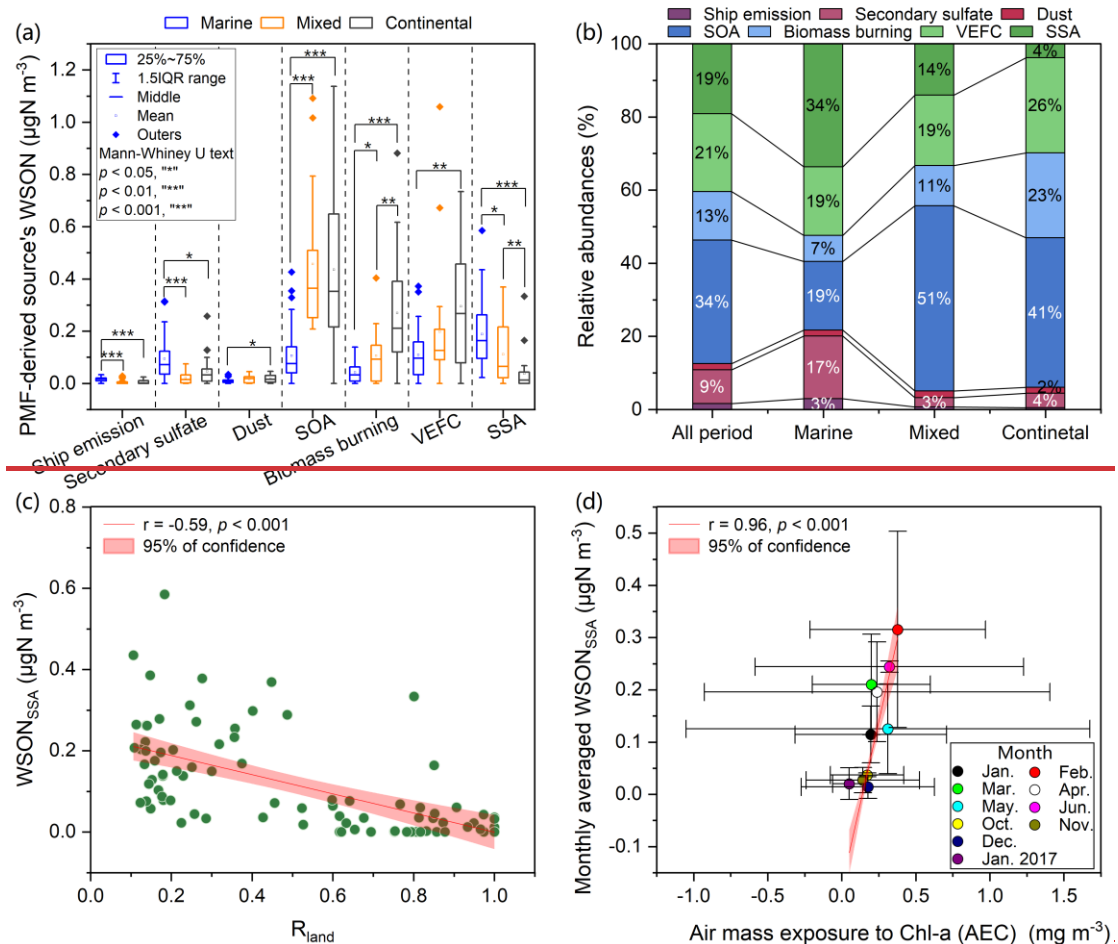
Over the entire study period, SOA ( $34\% \pm 25\%$ ), VEFC ( $21\% \pm 16\%$ ), SSA ( $19\% \pm 19\%$ ), and BB ( $13\% \pm 12\%$ ) emerged as the dominant sources of WSON in Bangkok aerosols (Figures 3b and S9). This is consistent with previous reports highlighting secondary formation and ~~biomass burning~~BB as major contributors to WSON (Leung *et al.*, 2024; Tsagkaraki *et al.*, 2021; Yu *et al.*, 2017). Earlier factor-based studies also indicated that sea salt can explain over 20% of the variance in WSON (Chen and Chen, 2010; Shi *et al.*, 2010). The contribution of SSA in our study, however, exceeded values reported for other coastal regions such as Hong Kong (4.4%) and the Eastern Mediterranean (<5%) (Leung *et al.*, 2024; Nehir and Koçak, 2018; Tsagkaraki *et al.*, 2021). Two factors may explain this discrepancy. First, ~~organic-nitrogen~~ON in coarse

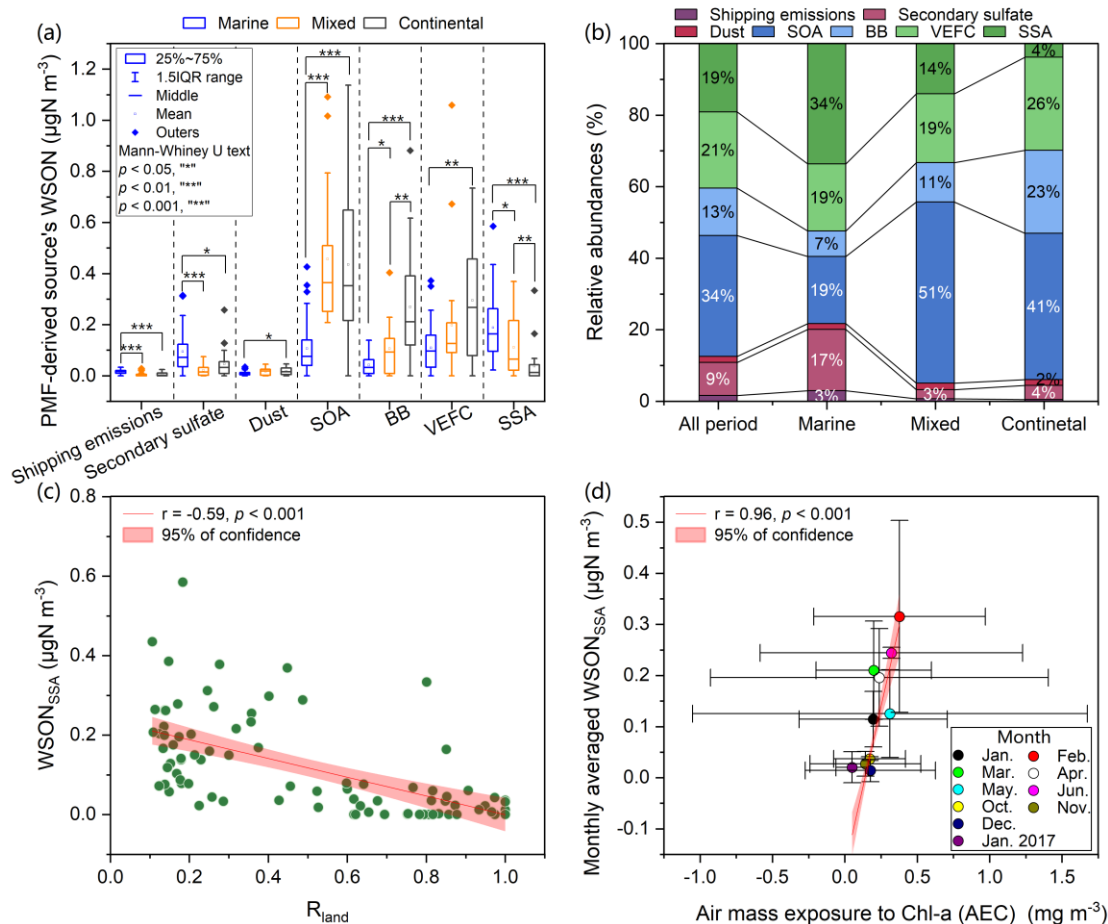
aerosols often originates from soil, dust, or large sea-salt particles (Cornell *et al.*, 2001; Mace *et al.*, 2003), whereas studies focusing on PM<sub>2.5</sub>—such as those in Hong Kong—naturally record lower sea-salt contributions (Leung *et al.*, 2024). Second, the ultra-oligotrophic marine environment of the Eastern Mediterranean, characterized by low nutrient availability and limited riverine input, results in low marine productivity and thus diminished marine-derived WSON (Nehir and Koçak, 2018; Tsagkaraki *et al.*, 2021).

We further disaggregated WSON source contributions by air mass regime (Figure 3b). Under marine influence, SSA constituted the dominant source of WSON (34% ± 17%), exceeding SOA (19% ± 17%), ~~and~~ VEFC (19% ± 14%), ~~and secondary sulfate (17% ± 16%)~~, while BB contributed minimally (7.1% ± 6.0%). This pattern is consistent with studies conducted in remote marine and island settings (Altieri *et al.*, 2016; Miyazaki *et al.*, 2011; Violaki *et al.*, 2015). Under mixed marine–continental influence, SOA became the dominant contributor (51% ± 20%), followed by VEFC (19% ± 14%) and SSA (14% ± 15%). During continental conditions, SOA remained the primary source (41% ± 26%), likely—~~reflecting multiple secondary formation pathways of nitrogen-containing organic aerosol, of which nitroaromatics may represent one possible subset of these compounds due to the formation of nitroaromatic compounds from biogenic and anthropogenic volatile organic compounds under NO<sub>x</sub> and H<sub>2</sub>O<sub>2</sub> exposure~~ (Xie *et al.*, 2017). Previous work has shown that oxidized  $\alpha$ -pinene SOA can account for 33%–~~37.78%~~ of WSON, with aerosol liquid water further promoting nighttime secondary WSON formation (Xu *et al.*, 2020). Under continental regimes, VEFC (26% ± 19%) and BB (23% ± 14%) also contributed substantially to WSON. Notably, the SSA contribution dropped sharply to 3.8% ± 6.4% under continental influence. Expressed relative to total WSTN, SSA-associated WSON contributed approximately 1.6% ± 2.1%, 7.3% ± 7.6%, and 13% ± 8.2% under continental-, mixed-, and marine-influenced conditions, respectively, with an overall mean contribution of 7.8% ± 8.2% over the sampled annual cycle, further illustrating its enhanced importance during marine influence.

Temporal variations in source-resolved WSON concentrations are shown in Figure 3a. Among the three air mass regimes, ~~SSA-derived~~ SSA-associated WSON concentrations peaked under marine influence (0.19 ± 0.12  $\mu\text{gN m}^{-3}$ ), approximately 1.7 times higher than during mixed periods (0.11 ± 0.12  $\mu\text{gN m}^{-3}$ ).

<sup>3</sup>) and five times higher than during continental periods ( $0.037 \pm 0.069 \mu\text{gN m}^{-3}$   
<sup>3</sup>). ~~Shipping-emission-derived-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 (~3%). WSON associated  
with the secondary sulfate factor under marine influence ( $0.094 \pm 0.086 \mu\text{gN}$   
 $\text{m}^{-3}$ ) 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  
~~underscore indicate that~~ marine air mass transport ~~as the principal driver of~~  
~~SSA-derived~~ 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-~~derived-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 ~~contributions concentrations in this dataset, suggesting limited~~  
This suggests that wet scavenging alone did not dominate the observed source-  
resolved influence on WSON variability composition.





**Figure 3.** Source apportionment of WSON based on PMF. (a) Absolute concentrations and (b) relative contributions of PMF-resolved sources to total WSON during the entire study period and under marine-, mixed-, and continental-influenced conditions. (c) Correlation between  $R_{\text{land}}$  and  $\text{SSA}$ -derived  $\text{SSA}$ -associated WSON. (d) Relationship between air mass exposure to chlorophyll-a (AEC) and monthly averaged  $\text{SSA}$ -derived  $\text{SSA}$ -associated WSON concentrations, color-coded by sampling month.

### 3.3. Marine Productivity as a Key Factor ~~influenced~~ influencing ~~coastal~~ Coastal WSON Distribution

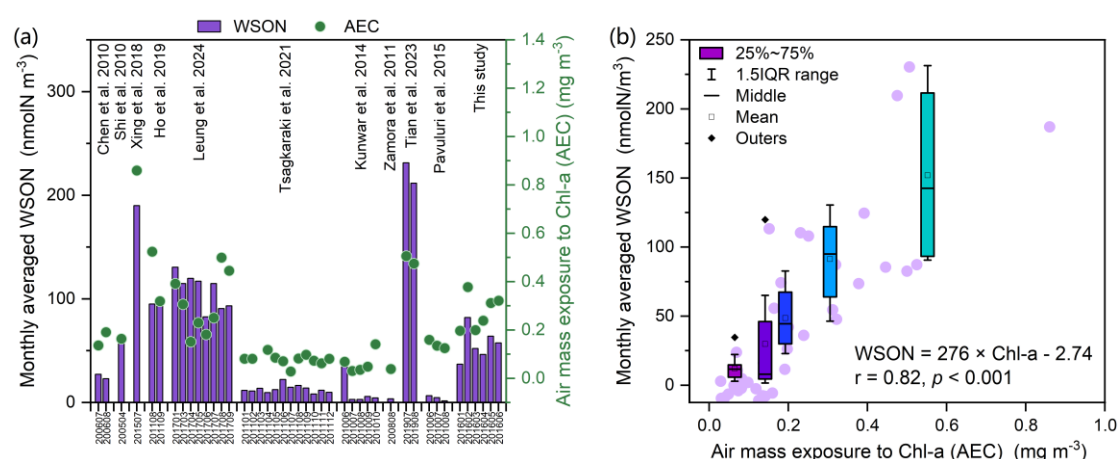
$\text{SSA}$  is dominated by inorganic sea salt but can also comprises an important complex mixture of organic compounds fraction derived from the ocean-ocean-surface materials (Prather *et al.*, 2013; Quinn *et al.*, 2014; Schiffer *et al.*, 2018). Numerous Previous studies have identified linked marine biological productivity to the organic enrichment of SSA as a major source of WSON in remote marine regions (O'Dowd *et al.*, 2015; Violaki *et al.*, 2015). Satellite-derived chlorophyll-a (and Chl-a) concentrations, which provide global coverage of oceanic phytoplankton biomass, are has often been widely used as

a broad proxy for the organic ocean-surface biological conditions fraction in SSA (Facchini *et al.*, 2008; O'Dowd *et al.*, 2004). Consistent with this, ON and OC concentrations in marine aerosols from highly productive waters are approximately twice those from oligotrophic regions (Miyazaki *et al.*, 2010). Stable carbon isotope analyses further indicate that marine-derived carbon can contribute 46%–72% of total aerosol carbon in productive oceanic areas (Miyazaki *et al.*, 2010). A seasonal pattern linking both Chl-a and water-insoluble OC has been documented (O'Dowd *et al.*, 2008). Given the substantial contribution of SSA to associated WSON to total WSON and WSTN during marine-influenced periods in our study, we investigated further examined whether marine productivity as a key driver of WSON distribution was related to its variability.

We calculated air mass exposure to chlorophyll-a (AEC) values based on monthly MODIS Chl-a data (4 km resolution) along 120-hour backward trajectories (see Methods). SSA-derived SSA-associated WSON exhibited a strong positive correlation with AEC ( $r = 0.96$ ,  $p < 0.001$ ; Figure 3d), consistent with a linkage between marine productivity and the variability of this factor. highlighting phytoplankton's role in aerosol composition. This contrasts with Tian *et al.* (2023), who observed significant WSON-AEC correlations only in summer, likely reflecting stronger due to dominant anthropogenic influences suppressing and/or weaker marine signatures signals during other seasons. While previous multivariate regression identified wind speed and Chl-a as key predictors of the organic fraction in SSA (Gantt *et al.*, 2011), other studies note that Chl-a alone may not fully capture organic enrichment (Rinaldi *et al.*, 2013). Nevertheless Still, moderate correlations ( $r \approx 0.60$ ) between Chl-a and marine organic aerosol abundance have been reported (Sciare *et al.*, 2009). Collectively, these findings indicate that biogenic organic matter enriched in lipids, proteins, and humic substances in the sea surface microlayer and transferred via bubble bursting is a primary source of WSON in Bangkok during marine air mass influence. 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.



To better understand the large-scale role of oceanic WSON sources in coastal regions affected by marine air masses, we further examine the broader relevance of this relationship, we compiled and analyzed a global dataset of WSON concentrations from coastal and island sites and recalculated air-mass trajectories. We consistently applied HYSPLIT trajectory analysis,  $R_{land}$  calculation, and AEC estimation values based on MODIS Chl-a to for all each sites (Figure 4a and Table S3). Across these marine-influenced coastal datasets, the highest WSON concentrations occurred at Huaniao Island (Tian *et al.*, 2023), followed by Jiaozhou Bay (Xing *et al.*, 2018), Hong Kong (Ho *et al.*, 2019; Leung *et al.*, 2024), the South China Sea (Shi *et al.*, 2010), and Bangkok. The lowest values were observed in the Eastern Mediterranean (Tsagkaraki *et al.*, 2021), Keelung City (Chen and Chen, 2010), Okinawa Island (Kunwar and Kawamura, 2014), Barbados (Zamora *et al.*, 2011), and Sapporo (Pavuluri *et al.*, 2015). Notably, spatial patterns in AEC closely mirrored those in WSON. A significant positive correlation was found between WSON and AEC across all sites ( $r = 0.82$ ,  $WSON [nmol m^{-3}] = 276 \times AEC [mg m^{-3}] - 2.74$ , Figure 4b), that WSON levels in marine-influenced coastal aerosols are strongly linked to ocean surface productivity along the air mass transport pathway. This large-scale comparison supports the broader consistency of the observed relationship, although the influence of site-to-site differences in sampling protocol, aerosol size fraction, and anthropogenic impact cannot be excluded.



**Figure 4.** (a) Geographic distributions and (b) correlation between monthly mean air mass exposure to Chl-a (AEC) concentrations values and monthly mean WSON concentration in coastal regions predominantly influenced by marine air masses. Values are provided in

Table S3. Air mass trajectories for these coastal and island sampling sites were recalculated using the HYSPLIT model, and AEC values were derived from MODIS monthly Chl-a concentrations.

#### 4. Conclusions

The relationship equation derived in this study ( $WSON [nmol\ m^{-3}] = 276 \times AEC [mg\ m^{-3}] - 2.74$ ) 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. ~~quantitative means to link marine aerosol composition with oceanic biological activity. In the context of rising sea surface temperatures, enhanced microalgal growth may elevate emissions of marine primary organic aerosol, thereby introducing an important component into the aerosol-cloud-climate feedback system. These findings highlight the necessity of incorporating marine-derived organic nitrogen into climate models to better represent interactions between marine ecosystems, atmospheric chemistry, and climate regulation. Variations in marine productivity whether natural or anthropogenically driven can directly modulate atmospheric nitrogen levels, which may in turn influence oceanic nitrogen deposition and primary productivity. Given that organic nitrogen deposition may also stimulate CO<sub>2</sub> uptake, this study underscores the role of marine ecosystems in climate mitigation and calls for a more integrated understanding of global climate dynamics.~~

~~Our results~~ 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. ~~reveal a previously underestimated source of atmospheric WSON derived from marine biota, particularly during periods of marine air mass influence. The strong correlation between WSON and chlorophyll-a based AEC offers new evidence for the role~~

~~of marine productivity in shaping atmospheric nitrogen composition. In contrast to earlier studies that have largely overlooked this pathway, our analysis identifies marine-derived organic nitrogen as a relevant contributor, especially in productive coastal regions. This omission in current climate models may lead to incomplete representations of nitrogen cycling and aerosol processes. We therefore propose suggest that integratingthe integration of satellite-derived Chl-aehlorophyll-a data into air mass trajectory analyses may help improve future assessments of marine-related WSON variability, coupled with the empirical relationship presented here, to better quantify marine-sourced WSON. Integrating this component into climate models will refine predictions of nitrogen deposition, ecosystem responses, and atmospheric composition.~~

#### **Code and data availability.**

Data are available from the link (<https://doi.org/10.17605/OSF.IO/YMJ3F>).

#### **Supplement**

The supplement related to this article is available online.

#### **Author contributions**

Conceptualization: JT. Funding acquisition: JT, SH, SZ, and GZ. Investigation: JT, XW, JW, SL, XG, GuZ, and SB. Methodology: JT, XW, and JW. Project administration: GZ and SZ. Resources: YM, SZ, JL, and GZ. Software: XW, and SL. Supervision: JL, SZ, and GZ. Validation: JT and SZ. Writing (original draft): JT. Writing (review and editing): XW, SH, SZ, and GZ.

#### **NotesCompeting interests**

The authors declare no competing financial interest.

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