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Marine-Derived Water-Soluble Organic Nitrogen in Coastal Air: Influence of Ocean Productivity on 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 (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 NO_3^- and 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 in water-soluble total nitrogen remained consistent, implying a persistent marine source. Positive matrix factorization revealed that the contribution of sea spray aerosol (SSA)-derived WSON increased markedly with oceanic influence, accounting for 3.8% ± 6.4%, 14% ± 14%, and 34% ± 17% under continental, mixed, and marine conditions, respectively. Moreover, marine productivity, assessed via air mass exposure to chlorophyll a concentrations (AEC), exhibited a strong positive correlation with SSA-derived WSON ($r = 0.96$, $p < 0.001$), a finding supported by large-scale reanalysis. These results provide direct evidence that marine organic aerosols represent a major source of WSON in coastal regions globally, with important implications for atmospheric nitrogen cycling and climate feedback processes.

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1. Introduction

Organic nitrogen (ON), which includes compounds such as amino acids, urea, organic nitrates, nitroaromatics, and humic 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 (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^{-1}, comparable in magnitude to anthropogenic emissions from fossil fuel combustion and biomass burning (Ito *et al.*, 2014; Kanakidou *et al.*, 2012). Consequently, organic nitrogen in marine aerosols is considered to be primarily derived from 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 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.*,

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2024; Tsagkaraki *et al.*, 2021). This discrepancy indicates that distinguishing anthropogenic from marine sources remains a challenge, even in coastal and adjacent marine environments.

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—the gradual enrichment of aquatic ecosystems with nutrients such as nitrogen—enhances primary productivity and 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^3 min^{-1}) 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), Wet (April–June and October 2016), and Dry II (November 2016–January 2017). Sampling frequency averaged 5 ± 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. 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 °C until analysis. A

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summary of TSP mass concentrations, chemical components, and meteorological conditions is provided in Table S1 (*Supplement*).

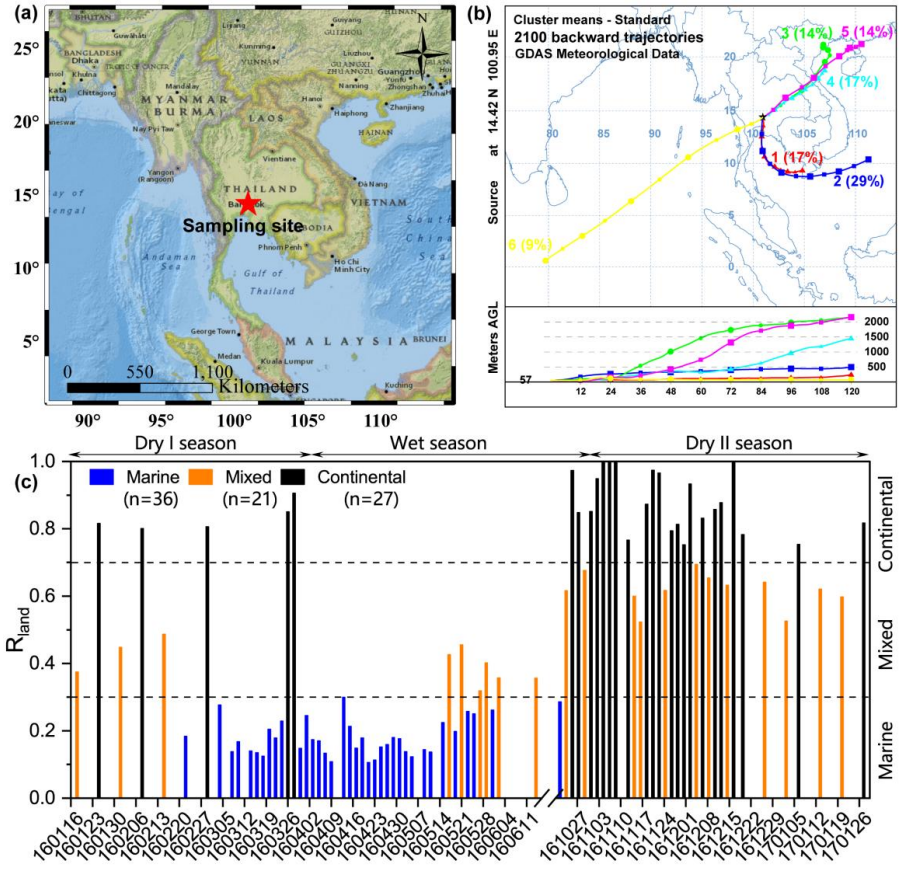

Figure 1. (a) Sampling site location in Bangkok, Thailand. (b) Classified air mass trajectories (detailed in Figure S1–S2). (c) Distribution of the air mass retention ratio over land (R_{land}), with samples categorized as marine-influenced ($R_{land} < 0.3$), mixed-influenced ($0.3 \leq R_{land} \leq 0.7$), 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 NIOSH870 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

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

Polar molecular tracers—including biomass burning markers (levoglucosan, mannosan, galactosan) and biogenic/anthropogenic secondary organic aerosol 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 ^{13}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 > 18.2M·cm^{-1}), followed by filtration through 0.22 μm PTFE membranes. Concentrations were measured with a total organic carbon/total nitrogen analyzer (model TOC-Vcsh, Shimadzu). WSON was calculated as the difference between WSTN and water-soluble inorganic nitrogen (WSIN), where WSIN comprises NH_4^+-N, NO_3^--N, NO_2^--N: [WSON] = [WSTN] − [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 species may not be fully converted to nitrogen monoxide in the TOC/TN analyzer, potentially leading to underestimation of WSON (Miyazaki *et al.*, 2011).

The relative standard deviation (RSD) for WSTN analysis were 3.6% (method) and 0.77% (instrument). Method detection limits were 0.09 μg m^{-3} for WSON, 0.03 μg m^{-3} for NO_3^-, and 0.02 μg m^{-3} for NH_4^+. Field blank levels were 0.067 μgN m^{-3} (WSON), 0.043 μgN m^{-3} (NH_4^+-N), and 0.07 μgN m^{-3} (NO_3^--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 WSON and WSTN concentrations were blank-corrected.

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2.3 Source Apportionment

The U.S. Environmental Protection Agency's Positive Matrix Factorization 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, WSON was included as a total variable to resolve its sources. Following the base run, rotational stability (F_{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.

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° × 1° 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 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/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.

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

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 ($R_{land} > 0.7$). This classification is further supported by molecular marker analysis: during marine-influenced periods, the regression slope for Na^+ versus Mg^{2+} (0.11) closely aligned with the seawater reference ratio (0.12). Elevated levels of Cl^-, Na^+, non-sea-salt sulfate (nss-SO_4^{2-}), and $Na^+/\sum$ions ratio consistently reflected dominant contributions from sea spray emissions. Although long-range anthropogenic transport may contribute to nss-SO_4^{2-}, marine biogenic sources prevailed, with concentrations significantly higher during marine-influenced periods than during mixed or continental periods. In contrast, combustion-derived species such as non-sea-salt K^+ (nss-K^+), EC, and levoglucosan showed markedly higher concentrations during continental- and mixed-influenced periods than during marine-influenced periods (Table S2).

Air mass exposure to chlorophyll-a (AEC), defined as the mean sea surface chlorophyll-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:

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

Here, N_{total} denotes the total number of hourly endpoints (120, including the receptor point) along the trajectory. The variable $Chla_i$ represents the mean chlorophyll-a concentration—derived from MODIS-Aqua monthly composites at a 4 km resolution—within a 20 km radius of the i_{th} trajectory

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endpoint. Endpoints over land were assigned $Chla_i$ = zero. The weighting factor $e^{-h_i/500}$ accounts for the influence of altitude h_i, reflecting the reduced contribution from higher altitudes due to chlorophyll dilution and particle deposition during transport. The denominator $n$corresponds to the number of trajectory endpoints with valid chlorophyll-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 ranged from 17 to 161 μg m^{-3} (mean: 55 ± 30 μg m^{-3}), 3.7 to 38 μg m^{-3} (mean: 12 ± 6.3 μg m^{-3}), and 0.16 to 2.8 μg m^{-3} (mean: 1.4 ± 0.43 μg m^{-3}), respectively. The TSP level in this region were substantially lower than those reported in other areas, such as the Eastern Mediterranean (219.90 ± 104.85 μg m^{-3}; Tripathee *et al.*, 2021), Jiaozhou Bay (133.7 ± 80.35 μg m^{-3}; Xing *et al.*, 2018), and Xi'an during the dust episodes (2109 ± 1360 μg 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. 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, $p < 0.001$), indicating efficient aerosol removal by

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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% ± 1.1% (range 2.2%–7.2%), higher than that reported in the Eastern Mediterranean (~ 2.7%; Tripathee *et al.*, 2021) and comparable to values from Sapporo, Japan (3.81% ± 2.28%; Pavuluri *et al.*, 2015), underscoring the relevance of water-soluble nitrogen in this region. As shown in Figure 2a and Table S2, concentrations of WSON, NO_3^--N, and NH_4^+-N ranged from 0.27–2.3 μgN m^{-3} (0.95 ± 0.40 μgN m^{-3}), below detection limitation (BDL)–2.7 μgN m^{-3} (0.60 ± 0.52 μgN m^{-3}), and BDL–2.2 μgN m^{-3} (0.47 ± 0.44 μgN m^{-3}), respectively. WSON correlated positively with both TSP ($r = 0.65$, $p < 0.01$) and WSIN ($r = 0.51$, $p < 0.01$), suggesting possible common sources or formation pathways (Figure S6).

Concentrations of WSON, NO_3^--N, 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 NO_3^--N levels were significantly lower during marine-influenced periods than under mixed or continental conditions (Figure 2c, $p < 0.001$). In contrast, NH_4^+-N concentrations were slightly elevated during marine periods, consistent with the ocean potentially acting as a source of atmospheric ammonia (Altieri *et al.*, 2014). This pattern aligns with earlier studies reporting higher WSON in aerosols of remote marine regions under continental influence than in those of purely oceanic origin, indicating a stronger terrestrial contribution (Jickells *et al.*, 2013). Biomass burning tracers (e.g., levoglucosan, galactosan, mannosan) were also significantly lower during marine-influenced days (see Table S2). Furthermore, WSON correlated with biomass burning markers and SOA markers and aerosol liquid water content (ALWC, Text S3) under mixed and continental conditions—associations absent 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 (Figure S5b).

The WSON/WSTN ratio during marine-influenced days (50% ± 17%) was similar to that under continental influence (48% ± 15%) but lower than during mixed conditions (60% ± 17%) (Figure 2d). Continental WSON/WSTN ratios likely reflect anthropogenic emissions, whereas marine periods may be

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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., nitrate, ammonium) are efficiently removed by rainfall (Matsumoto *et al.*, 2019b; Nehir and Koçak, 2018), as shown by their strong correlation with precipitation (Figure S5c,d), whereas the lack of such a correlation for WSON suggests distinct scavenging behavior.

Annually, WSON accounted for 52% ± 17% of the WSTN (Figure 2b)—substantially higher than values reported from a forest site (20% ± 11%; Miyazaki *et al.*, 2014), an offshore island (27%; Tian *et al.*, 2023), Sapporo (9.2% ± 7.3%; Pavuluri *et al.*, 2015), and coastal Qingdao (~20%; Shi *et al.*, 2010). Elevated WSON/WSTN ratios have been documented in source emissions such as biomass burning (80% ± 6.3%), vehicle exhaust (67% ± 16%), and ship emissions (54% ± 31%) (Yu *et al.*, 2017) , as well as in polluted continental regions like Hawaii (64%; Cornell *et al.*, 2001) and Xi'an (45%, range: 22–68%; Ho *et al.*, 2015). By comparison, the South China Sea—characterized by marine background conditions—exhibited a WSON/WSTN ratio of 34%, higher than that of the Yellow Sea (17%; Shi *et al.*, 2010). During phytoplankton blooms, organic nitrogen can dominate aerosol composition, contributing up to 84% of total dissolved nitrogen (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 influence or marine biogenic activity. Therefore, the WSON/WSTN ratio alone is insufficient to discriminate marine and anthropogenic sources of WSON. More quantitative approaches are needed to apportion the origins of aerosol organic nitrogen.

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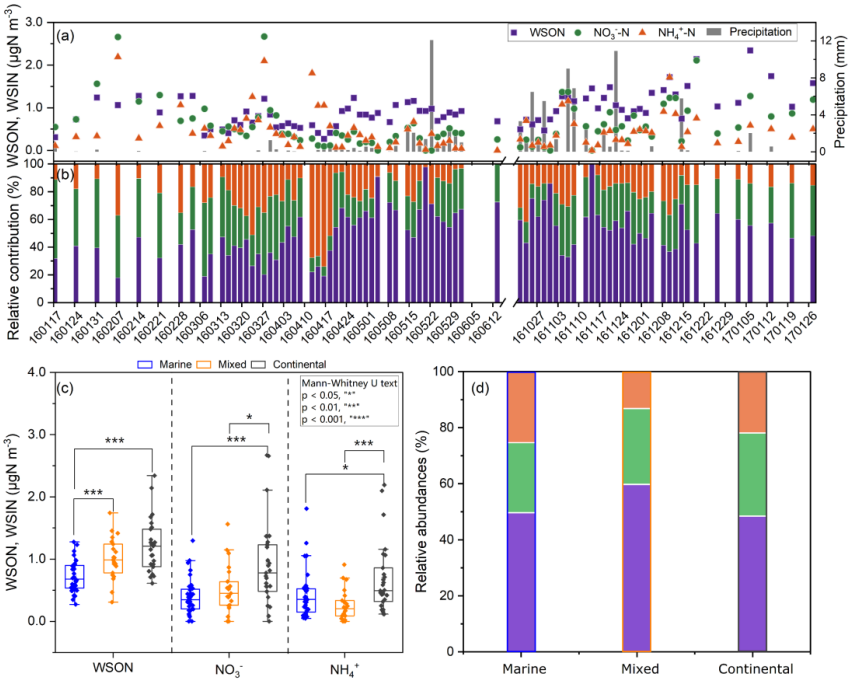

Figure 2. Temporal variations and source influences on nitrogen species in Bangkok aerosols. (a) Time-series of concentrations of WSON and WSIN (NH_4^+-N, NO_3^--N), overlaid with daily rainfall from ECMWF reanalysis data. (b) Relative contributions of WSON and WSIN to total water-soluble nitrogen across 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 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 emissions, secondary sulfate, dust, secondary organic aerosol (SOA), biomass burning (BB), vehicle emissions and fossil combustion (VEFC), and sea spray aerosol (SSA) (Figure S8). Here, the SSA factor was identified by high loadings of Na^+, Cl^-, and Mg, because we note that SSA-derived WSON was significantly associated with marine biological activity—not merely sea-salt particles—as elaborated below.

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Furthermore, PSCF mapping of SSA-derived WSON was mainly associated with the Gulf of Thailand and, to a lesser extent, the Bay of Bengal, reinforcing the role of sea spray as a relevant WSON source.

Over the entire study period, SOA (34% ± 25%), VEFC (21% ± 16%), SSA (19% ± 19%), and BB (13% ± 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 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 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 $PM_{2.5}$—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%), 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 due to the formation of nitroaromatic compounds from biogenic and anthropogenic volatile organic compounds under NO_X and H_2O_2 exposure (Xie *et al.*, 2017). Previous work has shown that oxidized *α*-pinene SOA can account for 33%–37.7% of WSON, with aerosol liquid water further promoting nighttime secondary WSON formation (Xu *et al.*, 2020). Under continental regimes, VEFC (26% ± 19%) and

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BB (23% ± 14%) also contributed substantially to WSON. Notably, the SSA contribution dropped sharply to 3.8% ± 6.4% under continental influence.

Temporal variations in source-resolved WSON concentrations are shown in Figure 3a. Among the three air mass regimes, SSA-derived WSON concentrations peaked under marine influence (0.19 ± 0.12 μgN m^{-3}), approximately 1.7 times higher than during mixed periods (0.11 ± 0.12 μgN m^{-3}) and five times higher than during continental periods (0.037 ± 0.069 μgN m^{-3}). Ship-emission-derived WSON was also elevated during marine days (0.015 ± 0.0075 μgN m^{-3}) relative to mixed and continental periods, though its overall contribution remained low (~3%). These results underscore marine air mass transport as the principal driver of SSA-derived WSON, further supported by a strong negative correlation between SSA-WSON and R_{land} (Figure 3c, $r = -0.59$, $p < 0.001$). In contrast, SOA-, BB-, and VEFC-derived 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 correlation was observed between precipitation and PMF-resolved source contributions, suggesting limited wet scavenging influence on WSON composition.

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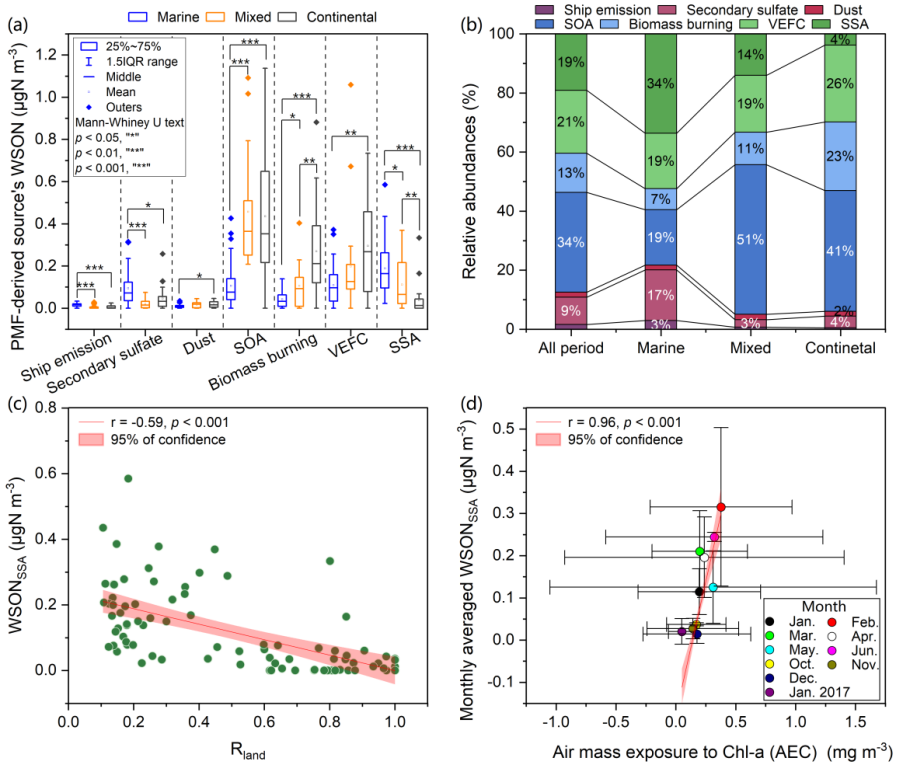

Figure 3. Source apportionment of WSON based on PMF. (a) Absolute concentrations and (b) relative contributions of PMF-resolved sources to total WSON during the entire study period and under marine-, mixed-, and continental-influenced conditions. (c) Correlation between R_{land} and SSA-derived WSON.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

3.3. Marine Productivity as a Key Factor influenced coastal WSON Distribution

SSA comprises a complex mixture of organic compounds derived from the ocean surface (Prather *et al.*, 2013; Quinn *et al.*, 2014; Schiffer *et al.*, 2018). Numerous studies have identified marine biological productivity as a major source of WSON in remote marine regions (O'Dowd *et al.*, 2015; Violaki *et al.*, 2015). Satellite-derived chlorophyll-a (Chl-a) concentrations, which provide global coverage of oceanic phytoplankton biomass, are widely used as a proxy for the organic fraction in SSA (Facchini *et al.*, 2008; O'Dowd *et al.*, 2004).

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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 WSON during marine-influenced periods in our study, we investigated marine productivity as a key driver of WSON distribution.

We calculated air mass exposure to chlorophyll-a (AEC) based on monthly MODIS Chl-a data (4 km resolution) along 120-hour backward trajectories (see Methods). SSA-derived WSON exhibited a strong positive correlation with AEC ($r = 0.96$, $p < 0.001$; Figure 3d), highlighting phytoplankton's role in aerosol composition. This contrasts with Tian *et al.* (2023), who observed significant WSON-AEC correlations only in summer, likely due to dominant anthropogenic influences suppressing marine signatures 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, 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.

To better understand the large-scale role of oceanic WSON sources in coastal regions affected by marine air masses, we compiled and analyzed a global dataset of WSON concentrations from coastal and island sites. We consistently applied HYSPLIT trajectory analysis, R_{land} calculation, and AEC estimation based on MODIS Chl-a to all sites (Figure 4a and Table S3). Spatially, 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).

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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 ×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.

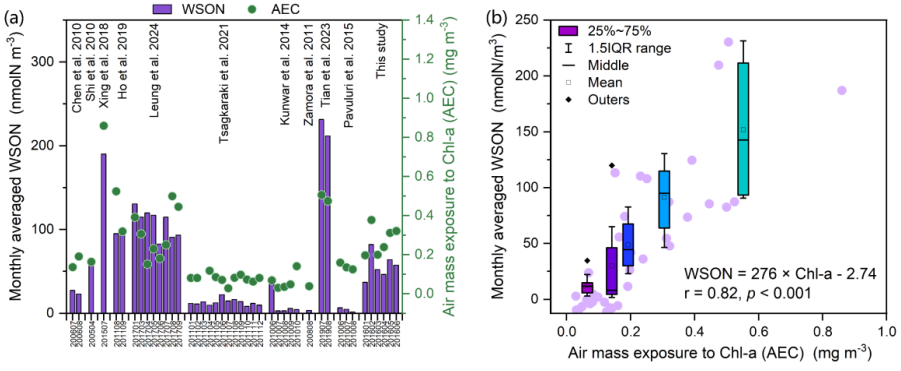

Figure 4. (a) Geographic distributions and (b) correlation between monthly mean air mass exposure to Chl-a (AEC) concentrations and monthly mean WSON concentration in coastal regions predominantly influenced by marine air masses. Values are provided in Table S3. Air mass trajectories for these coastal 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 derived in this study (WSON [nmol m^{-3}] = 276 ×AEC [mg m^{-3}] – 2.74) provides a 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_2

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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 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 the integration of satellite-derived chlorophyll-a data into air mass trajectory analyses, 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.

Notes

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