

Response Letter

Dear Editor,

Thank you very much for providing us this opportunity to revise our manuscript. We have carefully and thoughtfully addressed the referee's concerns. In the revised manuscript, we corrected the internal inconsistencies in the reported particle-phase water-soluble organic nitrogen (WSON) concentrations and the NO_3^- standard deviation in Table 1. We added basin meteorology and emission characteristics to justify the site selection. We also provided an in-depth discussion on amine partitioning mechanisms by treating methylamine (MA) and dimethylamine (DMA) as distinct species, incorporating species-specific parameters such as pK_a , Henry's law constant, and reaction rates, and distinguishing physical dissolution from chemical-enhanced partitioning. In addition, we replaced the correlation heatmap with scatter plots to address both linear and nonlinear relationships, deleted unsupported statements on photochemistry and WSON diurnal variation, added Table 2 comparing F_{WSON} across three Chinese cities (Chengdu (CD), Shanghai (SH), and Xianghe (XH)), and standardized abbreviations, units, and author metadata throughout the manuscript. By addressing these concerns, we believe the scientific quality of the current version of the manuscript is improved a lot. Please see our detailed response below.

Sincerely yours

Gehui Wang

Sept. 14, 2026

1 **Referee: #1**

2 **General comments:**

3 This study simultaneously measured gas- and particle- phase water- soluble
4 organic nitrogen, using an online gas and aerosol composition monitoring system, ion
5 chromatography, and a total organic carbon/total nitrogen analyzer. By combining the
6 thermodynamic model, gas- particle partitioning coefficients, and sigmoidal curve
7 analysis, the authors investigated the gas- particle partitioning characteristics of
8 WSON and amines during different pollution stages and evaluated the effects of
9 aerosol liquid water content, particle acidity, and temperature. The overall research
10 framework is relatively complete. The topic is relevant to current research interests in
11 atmospheric chemistry and aerosol science, and the observational dataset has potential
12 value for publication. I therefore recommend acceptance after minor revision.

13 **Reply:** We thank you very much for your comments above and carefully revised the
14 manuscript according to the comments from you and another reviewer. See our
15 detailed responses below.

16 **Detailed comments:**

17 **Comments:**

18 1. The reported particle-phase WSON concentrations are internally inconsistent
19 The abstract and Table 1 report an average particle- phase WSON concentration of $2.3 \pm 1.4 \mu\text{gN m}^{-3}$, whereas Line 192 of the main text reports a value of $2.7 \pm 1.6 \mu\text{g m}^{-3}$.
20 $\pm 1.4 \mu\text{gN m}^{-3}$, whereas Line 192 of the main text reports a value of $2.7 \pm 1.6 \mu\text{g m}^{-3}$.
21 Line 200 again uses $2.3 \pm 1.4 \mu\text{gN m}^{-3}$. The authors should re-examine the original
22 dataset, the number of valid observations, and the treatment of missing values, and
23 should ensure that the values reported in the abstract, main text, Table 1, figure
24 captions, and Supplementary Information are fully consistent.

25 **Reply:** We sincerely thank the reviewer for identifying this internal inconsistency in
26 the reported particle-phase WSON concentrations. We have carefully re-examined our
27 original dataset, the number of valid observations, and our treatment of missing values.
28 Our verification confirmed that the value $2.3 \pm 1.4 \mu\text{gN m}^{-3}$ is the correct statistical
29 result derived from all valid observations. The value $2.7 \pm 1.6 \mu\text{gN m}^{-3}$ reported in

Line 192 of the original manuscript was an erroneous value from an intermediate calculation step. We have now corrected this to $2.3 \pm 1.4 \mu\text{gN m}^{-3}$ and systematically verified the consistency of this value throughout the abstract, main text (including the revised Line 215), Table 1, all figure captions, and the Supplementary Information. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 2, line 22-24):

“The average concentration of particulate WSON ($2.3 \pm 1.4 \mu\text{gN m}^{-3}$) was about twice that of gaseous WSON ($1.2 \pm 0.9 \mu\text{g N m}^{-3}$) and increased significantly as $\text{PM}_{2.5}$ increased.”

Revised text as it appears in the text (Page 9, line 215-218):

“The concentration of WSON_p ($2.3 \pm 1.4 \mu\text{gN m}^{-3}$) was more than twice that of WSON_g ($1.2 \pm 0.9 \mu\text{gN m}^{-3}$) (Fig. 1c and Table 1), indicating that WSON in CD mainly exists in the particle phase, as observed at Mt. Fuji in Japan (Matsumoto et al., 2020).”

Revised text as it appears in the text (Page 9, line 222-228):

“Our study shows that the wintertime WSON_p concentration of $\text{PM}_{2.5}$ in CD ($2.3 \pm 1.4 \mu\text{gN m}^{-3}$) was slightly higher than those in central Chinese cities such as Nanjing ($1.7 \pm 0.6 \mu\text{gN m}^{-3}$) (Guan et al., 2022) and Shanghai (SH, $1.9 \pm 1.8 \mu\text{gN m}^{-3}$) (Lv et al., 2022), as well as in the northern city of Wuhai ($1.7 \pm 1.1 \mu\text{gN m}^{-3}$) (Xie et al., 2025), but substantially higher than those in southern coastal cities such as Guangzhou ($0.48 - 0.53 \mu\text{gN m}^{-3}$) (Yu et al., 2017) and Hong Kong ($0.6 \pm 0.6 \mu\text{gN m}^{-3}$) (Leung et al., 2024).”

Revised text as it appears in the text (Page 24, line 718-721):

Table R1. Concentrations of major pollutants and meteorological parameters in Chengdu (CD) during winter.

Specie	Whole	Clean Period	Transition Period	Pollution Period
I. Gas pollutants ($\mu\text{g m}^{-3}$)				
NH ₃	12.3 ± 3.6 (2.4–31.2)	8.1 ± 2.7 (2.9–11.5)	11.8 ± 3.5 (3.4–31.2)	13.5 ± 3.0 (5.1–21.6)
WSO _{Ng} ^a	1.2 ± 0.9 (0.1–5.8)	0.7 ± 0.5 (0.1–1.7)	1.0 ± 0.7 (0.1–3.9)	1.4 ± 1.1 (0.1–5.8)
WSO _{Cg} ^b	22.6 ± 9.8 (4.4–69.8)	16.6 ± 7.9 (4.4–31.3)	20.3 ± 8.8 (4.8–51.5)	25.5 ± 9.8 (6.2–69.8)
CH ₃ NH ₂	1.23 ± 0.36 (0.46–2.90)	0.84 ± 0.21 (0.46–1.26)	1.19 ± 0.37 (0.46–2.90)	1.35 ± 0.32 (0.59–2.32)
(CH ₃) ₂ NH	0.41 ± 0.03 (0.34–0.68)	0.38 ± 0.02 (0.34–0.42)	0.40 ± 0.03 (0.34–0.68)	0.42 ± 0.03 (0.35–0.53)
II. Major components of PM _{2.5} ($\mu\text{g m}^{-3}$)				
PM _{2.5}	76 ± 38 (7–199)	29 ± 11 (7–49)	55 ± 18 (9–101)	100 ± 33 (30–199)
SO ₄ ²⁻	4.9 ± 2.0 (1.5–11.9)	2.9 ± 1.2 (1.5–6.0)	3.7 ± 1.0 (1.6–6.7)	6.2 ± 1.7 (2.9–11.9)
NO ₃ ⁻	16.7 ± 8.3 (2.9–39.0)	6.4 ± 2.1 (2.9–11.0)	12.0 ± 4.0 (3.4–22.3)	22.3 ± 7.2 (6.4–39.0)
Cl ⁻	1.3 ± 1.0 (0.1–6.1)	0.5 ± 0.2 (0.1–0.9)	1.0 ± 0.7 (0.1–6.0)	1.8 ± 1.1 (0.3–6.1)
NH ₄ ⁺	7.2 ± 3.0 (1.4–15.5)	3.5 ± 0.8 (2.1–5.5)	5.6 ± 1.7 (1.4–10.3)	9.1 ± 2.6 (3.0–15.5)
Other ions ^c	2.2 ± 1.1 (0.8–9.1)	1.2 ± 0.2 (0.8–1.9)	1.7 ± 0.5 (0.9–3.5)	2.8 ± 1.2 (1.1–9.1)
OM ^d	21.1 ± 9.6 (6.7–47.4)	9.5 ± 1.9 (6.7–11.3)	15.1 ± 3.5 (6.9–21.0)	25.6 ± 9.3 (8.1–47.4)
WSO _{Np} ^a	2.3 ± 1.4 (0.1–6.3)	1.0 ± 0.9 (0.1–4.3)	1.5 ± 0.8 (0.1–4.9)	3.0 ± 1.4 (0.3–6.3)
WSO _{Cp} ^b	13.5 ± 7.2 (1.2–56.2)	8.1 ± 4.5 (1.4–21.0)	10.9 ± 6.6 (1.2–56.2)	16.3 ± 6.8 (2.7–54.8)
CH ₃ NH ₃ ⁺	0.45 ± 0.24 (0.03–1.27)	0.18 ± 0.06 (0.10–0.34)	0.33 ± 0.13 (0.03–0.67)	0.61 ± 0.23 (0.15–1.27)
(CH ₃) ₂ NH ₂ ⁺	0.09 ± 0.05 (0.02–0.34)	0.06 ± 0.01 (0.04–0.09)	0.07 ± 0.04 (0.02–0.29)	0.12 ± 0.04 (0.05–0.34)
III. Gas-particle partitioning coefficients of WSON, WSOC, ammonium (AM), methylamine (MA), dimethylamine (DMA)				
F _{WSON} ^e	0.70 ± 0.14 (0.28–0.98)	0.62 ± 0.17 (0.29–0.88)	0.66 ± 0.14 (0.28–0.90)	0.73 ± 0.13 (0.42–0.98)
F _{WSOC} ^e	0.36 ± 0.09 (0.10–0.81)	0.32 ± 0.10 (0.19–0.65)	0.34 ± 0.09 (0.13–0.75)	0.39 ± 0.09 (0.10–0.81)
F _{AM} ^e	0.36 ± 0.08 (0.14–0.52)	0.31 ± 0.10 (0.19–0.52)	0.32 ± 0.07 (0.14–0.49)	0.39 ± 0.05 (0.20–0.49)
F _{MA} ^e	0.25 ± 0.08 (0.07–0.44)	0.18 ± 0.06 (0.09–0.31)	0.21 ± 0.06 (0.07–0.39)	0.30 ± 0.06 (0.12–0.40)
F _{DMA} ^e	0.18 ± 0.06 (0.04–0.45)	0.13 ± 0.02 (0.10–0.19)	0.15 ± 0.06 (0.04–0.42)	0.21 ± 0.05 (0.12–0.45)
IV. Meteorology parameters and aerosol properties				
T (°C)	6.4 ± 3.6 (-2.4–16.5)	6.2 ± 2.7 (0.2–12.3)	6.4 ± 3.4 (-1.3–15.4)	6.4 ± 3.9 (-2.4–16.5)
RH (%)	78 ± 19 (20–100)	78 ± 22 (26–99)	76 ± 19 (20–100)	79 ± 18 (29–100)
WS (m/s)	0.5 ± 0.4 (0.0–1.7)	0.6 ± 0.3 (0.0–1.4)	0.6 ± 0.4 (0.0–1.7)	0.3 ± 0.3 (0.0–0.9)
pH of PM _{2.5}	4.4 ± 0.2 (3.4–5.0)	4.2 ± 0.3 (3.4–4.5)	4.4 ± 0.2 (3.6–4.9)	4.4 ± 0.2 (3.8–5.0)
ALWC ^f	64.0 ± 71.5 (0.5–422.2)	34.0 ± 35.8 (1.2–130.5)	44.4 ± 49.2 (0.5–215.9)	87.6 ± 85.1 (1.6–422.2)

^a WSON_g is the gas-phase organics, WSON_p is the organics in PM_{2.5}, ($\mu\text{gN m}^{-3}$). ^b

WSOC_g is the gas-phase organics, WSOC_p is the organics in PM_{2.5}, ($\mu\text{gC m}^{-3}$). ^c

Other ions are the sum of NO₂⁻, F⁻, K⁺, Na⁺, Mg²⁺, and Ca²⁺. ^d Organic matter: OM =

OC × 1.6. ^e $F_p = C_p / (C_g + C_p)$. ^f ALWC: aerosol liquid water content of PM_{2.5}.

(Table R1 presents in the manuscript as Table 1)”

Comments:

2. The standard deviation and concentration range of nitrate during the clean period in Table 1 are clearly incompatible

Table 1 reports a NO₃⁻ concentration of 6.4 ± 32.1 $\mu\text{g m}^{-3}$ during the clean period, with a concentration range of 2.9–11.0 $\mu\text{g m}^{-3}$. A standard deviation of 32.1 $\mu\text{g m}^{-3}$ is mathematically impossible within this range. This value is likely a typographical or

calculation error, possibly 3.2 or 3.1 $\mu\text{g m}^{-3}$? The authors should thoroughly verify all values in Table 1, with particular attention to whether the data for the polluted and clean periods have been placed in the wrong columns and whether the units and significant figures are consistently reported.

Reply: We sincerely thank the reviewer for the careful reading and keen identification of this error. This was indeed a typographical mistake. The correct value should be $6.4 \pm 2.1 \mu\text{g m}^{-3}$, rather than $6.4 \pm 32.1 \mu\text{g m}^{-3}$ as originally reported. This error arose because the author inadvertently added an extra digit “3” when inputting the standard deviation; the intended value was “2.1” but was mistakenly entered as “32.1”. We have corrected the errors in Table 1 of the revised manuscript (see Table R1), and the changes have been highlighted in blue.

Regarding the units in Table 1, all values are reported in $\mu\text{g m}^{-3}$ unless otherwise specified. For species requiring special units (e.g., WSON, reported as $\mu\text{gN m}^{-3}$), this is clearly indicated by superscript annotations provided in the footnotes below the table.

Regarding the retention of significant figures, we followed common practice: $\text{PM}_{2.5}$ concentrations are reported as integers with no decimal places, inorganic ions are reported to one decimal place, and the gas-particle partitioning coefficients (F_p) are reported to two decimal places.

All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Comments:

3. The matching of datasets with different time resolutions and the effects of temporal autocorrelation should be incorporated into the statistical analysis. The temporal resolution of the WSON measurements was 3 h, whereas the temporal resolutions of the other ions, amines, and meteorological parameters were 1 h. The manuscript does not specify whether the hourly data were averaged over the corresponding 3 h WSON sampling intervals or whether the WSON data were interpolated to an hourly resolution.

Reply: We thank you for highlighting this critical methodological concern. We fully agree that the matching approach for datasets with different time resolutions should be explicitly stated in the statistical analysis. Following your suggestion, we have added the following clarification in **Section 2.2 (“Measurements of the gas- and aerosol-phase species”)** of the revised manuscript: “when calculating the WSIN concentration, the average is taken over the corresponding 3 h WSON sampling interval to match the time resolution of WSON.” Specifically, all hourly measurements (including inorganic ions, amines, and meteorological parameters) were arithmetically averaged over 3-hour windows to align with the 3-hour sampling interval of WSON. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 6, line 136-139):

“where, *g* and *p* represent the gas phase and particle phase, respectively; WSTN represents total water-soluble nitrogen measured by TOC/TN analyzer; WSIN represents the total water-soluble inorganic nitrogen measured by IGAC-IC, when calculating this value, the average is taken over a 3 h WSON sampling intervals.”

Comments:

4. Lines 320–327 state that amines are almost completely protonated at aerosol liquid water pH values of 0–5 and are therefore insensitive to changes in pH. This explanation conflates two distinct concepts: the fraction of dissolved amines that is protonated in the aqueous phase and the overall gas-particle partitioning fraction. Even when dissolved amines are predominantly protonated, their effective Henry’s law constants and total gas-particle partitioning may remain strongly dependent on pH. Ge et al. (2011) explicitly discussed the coupled dependence of amine partitioning on particle acidity, activity coefficients, and aerosol water content. The potential influence of pH therefore cannot be excluded solely on the basis of the dominant protonation state. DOI: 10.1016/j.atmosenv.2010.10.013. Pankow (2015) similarly emphasized that amine protonation, aerosol phase state, and volatility should be

treated within a unified phase-equilibrium framework. DOI:
10.1016/j.atmosenv.2015.09.056.

Based on the sigmoidal curve analysis, the manuscript concludes that “pH and temperature exert relatively weak effects on amine partitioning, whereas ALWC is the dominant factor.”

However, the observed PM_{2.5} pH range was only 3.4–5.0, as shown in Table 1. The sigmoidal curves in Figure 5 indicate that the protonation plateaus of MA and DMA, where ε is approximately 100%, extend to approximately pH 5–6, after which the protonated fractions begin to decline sharply.

Thus, nearly the entire observational dataset falls within the flat plateau region of the sigmoidal curves and does not cover the pH-sensitive transition region, approximately pH 6–8. The authors should reinterpret Figure 5 and revise the conclusion as follows: within the observed pH range and the uncertainty of the available measurements, no significant independent pH dependence was detected. The current dataset does not support the broader mechanistic conclusion that pH is intrinsically unimportant for amine partitioning.

Reply: Suggestion taken. We thank you for raising this important conceptual issue. We agree with your analysis that: (1) the protonation fraction of dissolved amines and the overall gas-particle partitioning coefficient are two distinct concepts; (2) even when dissolved amines are predominantly protonated, their effective Henry's law constants and total partitioning coefficients may still be strongly dependent on pH; and (3) the potential influence of pH cannot be excluded solely on the basis of the dominant protonation state.

The partitioning behavior of amines in the atmosphere is illustrated in Fig. R1. Once gaseous amines enter the particle phase, they are rapidly protonated to form $R_n\text{-NH}_{4-n}^+$, which subsequently form stable clusters with acidic species such as inorganic salts (sulfate and nitrate) (Schobesberger et al., 2013; Yao et al., 2018). Free unprotonated amines ($R_n\text{-NH}_{3-n}$ (aq)) are virtually absent in the particle phase, and thus the role of Henry's law in governing the partitioning between $R_n\text{-NH}_{3-n}$ (g) and

$R_n\text{-NH}_{3-n}$ (aq) is minimal. Tilgner et al. (2021) similarly stated in their original work: **“Since atmospheric waters are typically acidic, bases are predominantly present in their protonated form, and their partitioning is not greatly altered by typical variations in pH.”** Therefore, it is reasonable to use protonation to explain that amine partitioning in the atmosphere is weakly affected by pH variations.

Nevertheless, to ensure the rigor and scientific validity of our study, and to better illustrate the linear or nonlinear relationship between the overall gas-particle partitioning coefficient (F_p) and pH, we have replotted the scatter plot of their relationship, as shown in Fig. R2. It can be seen that there is no significant linear or nonlinear relationship between F_p and pH. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 13, line 358-366):

“To further identify the key factors governing the partitioning of amines, we analyzed the correlations between F_p and meteorological conditions (i.e., RH and T) as well as pH. As shown in Figures S6a–f, no significant linear or nonlinear correlations were observed between F_{MA} and T, RH, or pH during either daytime or nighttime ($R < 0.5$). Similarly, F_{DMA} did not show notable correlations with T, RH, or pH during the daytime ($R < 0.5$, $p > 0.05$). However, during the nighttime, F_{DMA} exhibited a significant nonlinear relationship with RH, with an exponential increase when RH exceeded 80%, while the influences of T and pH remained relatively weak ($R < 0.5$, $p > 0.05$).”

Revised text as it appears in the text (Page 14, line 375-379):

“However, given the acidity of atmospheric aqueous phases (pH 0–5), amines exist almost entirely in their protonated form ($R\text{-NH}_3^+$), within the observed pH range and the uncertainty of the available measurements, no significant independent pH dependence was detected (Tilgner et al., 2021).”

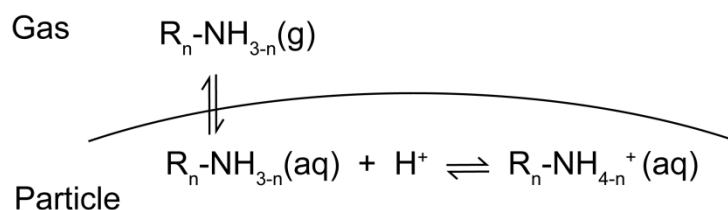


Figure R1. Schematic diagram of gas-particle distribution and electro-dissociation of amines.

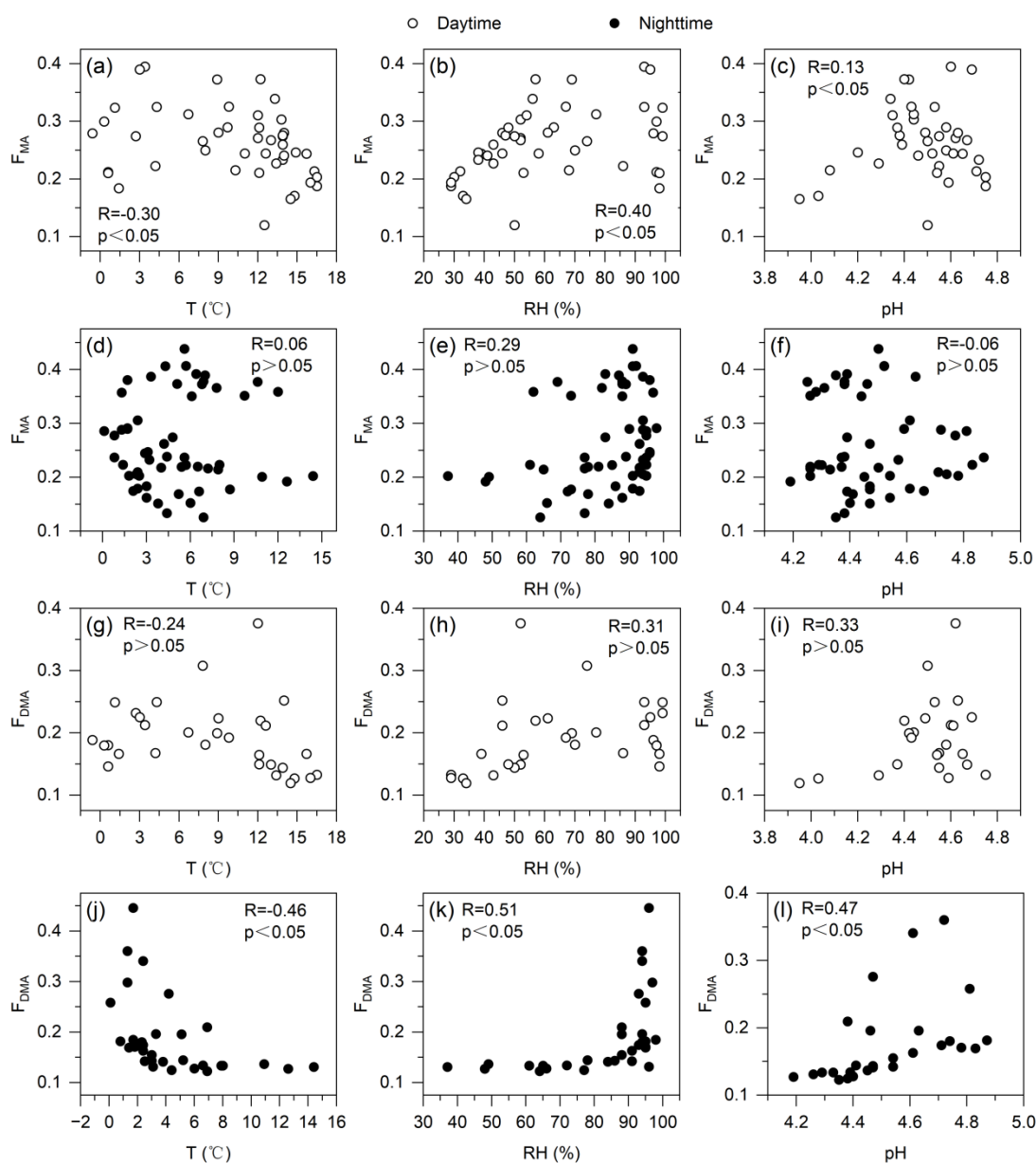


Figure R2. Factors affecting the gas-to-aerosol phase-partitioning coefficients of methylamine (F_{MA}) and dimethylamine (F_{DMA}) in Chengdu (CD) under the daytime and nighttime during the pollution period (2025.1.2—2025.1.6). (Figure R2 presents in the manuscript as Figure S6)

Comments:

5. Inappropriate citation regarding the volatility of organic nitrates

The citation of Pleim and Ran (2011) in Lines 53–55 is not appropriate for the discussion of the volatility of monofunctional and multifunctional organic nitrates. That paper primarily addresses surface fluxes in air quality models rather than the gas-particle partitioning of organic nitrates. DOI: 10.3390/atmos2030271.

The authors should replace this reference with studies that directly investigate the volatility, phase partitioning, or effective saturation concentrations of organic nitrates.

Reply: Thank you very much for pointing out the incorrect citation.

Rollins et al. (2013) observed that approximately 21% of alkyl and polyfunctional organic nitrates exist in the condensed phase, which indirectly confirms that polyfunctional nitrates constitute an important component of the particulate phase. González-Sánchez et al. (2021) noted that although alkyl and alkene nitrates are highly volatile, polyfunctional organic nitrates may exhibit much lower volatility and can partition into condensed phases such as aqueous and aerosol phases. Based on the findings reported in these two studies, we have corrected the literature citations in the manuscript. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 3, line 44-49):

“For example, low-molecular-weight amines and nitroaromatic compounds are semi-volatile and partition dynamically between phases (Liu et al., 2018; Chen et al., 2025), while monofunctional organic nitrates predominantly reside in the gas phase, whereas multifunctional ones preferentially partition into the particle phase (Rollins et al., 2013; González-Sánchez et al., 2021).”

Comments:

6. Incorrect publication year for the IMPROVE_A paper by Chow et al.

For the IMPROVE_A paper by Chow et al., Volume 57, pages 1014–1023 correspond

to the formal publication year of 2007, whereas the manuscript lists the year as 2012. Both PubMed and the journal record identify the publication information as 2007. DOI: 10.3155/1047-3289.57.9.1014. The authors should verify and correct the publication year in both the main text and the reference list.

Reply: We thank you for pointing out this error in the publication year. We have verified this reference and confirmed that the IMPROVE_A temperature protocol paper by Chow et al. was formally published in 2007 (Volume 57, pages 1014–1023), rather than 2012 as originally cited. We have corrected the publication year to 2007 in both the in-text citation and the reference list of the revised manuscript, with the changes highlighted in blue. We appreciate your rigorous review. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 6, line 135-138):

“The organic carbon (OC) and elemental carbon (EC) of the PM_{2.5} filter samples were analyzed by DRI Model 2015 Carbon Analyzer (Atmoslytic, Inc., Calabasas, USA) with IMPROVE_A protocol (Chow et al., 2007; Li et al., 2025).”

Reference:

“Chow, J. C., Watson, J. G., Chen, L. W. A., Chang, M. C. O., Robinson, N. F., Trimble, D., and Kohl, S.: The IMPROVE_A Temperature Protocol for Thermal/Optical Carbon Analysis: Maintaining Consistency with a Long-Term Database, J. Air Waste Manage. Assoc., 57, 1014-1023, <https://doi.org/10.3155/1047-3289.57.9.1014>, 2007.”

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1 **Referee: #2**

2 **General comment:**

3 This manuscript presents simultaneous measurements of gaseous and particulate
4 water-soluble organic nitrogen (WSO_N) in urban Chengdu during winter,
5 investigating the gas-particle partitioning coefficients (F_p), their diurnal variations,
6 and the dominant role of aerosol liquid water content (ALWC) driven by NH₄NO₃.
7 While the topic aligns with the scope of the journal Atmospheric Chemistry and
8 Physics (ACP) and the data acquisition appears rigorous, the current version lacks the
9 mechanistic depth expected for a process-oriented study. I recommend publication
10 after a Major Revision addressing the conceptual and methodological concerns
11 outlined below.

12 **Reply:** We thank you very much for your comments above and carefully revised the
13 manuscript according to the comments from you and another reviewer. See our
14 detailed responses below.

15 **Detailed comments:**

16 My major concern is regarding about the overlap with the authors' prior work. The
17 analytical framework employed here is highly consistent with the authors' previous
18 studies on gas-particle partitioning in different regions. The conclusion that ALWC
19 dominates WSO_N partitioning while pH and temperature are relatively weak largely
20 replicates the findings of the aforementioned studies. Chengdu is not merely as
21 another high-humidity site comparable to other areas. So I strongly recommend the
22 authors to refocus the discussion on how the unique meteorological and topographic
23 constraints of Chengdu modulate WSO_N partitioning behaviors differently than those
24 reported in coastal or northern China cities.

25 **Reply:** We sincerely thank you for this important concern regarding the overlap with
26 our prior work. We fully agree that Chengdu (CD) should not be treated simply as
27 another high-humidity site. In the revised manuscript, we have refocused the
28 discussion to highlight how the unique topographic and meteorological constraints of
29 CD modulate WSO_N partitioning differently from coastal and northern Chinese cities.

Specifically, we have added Table 2 comparing $F_{\text{WSO}}N$ across CD (representing the Sichuan Basin, SCB), Shanghai (SH, representing the Yangtze River Delta, YRD), and Xianghe (XH, representing the North China Plain, NCP). $F_{\text{WSO}}N$ was highest in CD (0.70 ± 0.14), followed by SH (0.49 ± 0.26), and lowest in XH (0.43 ± 0.23). This regional variation did not follow the trends of T and RH, and no significant differences in pH were observed, suggesting that T, RH, and pH exerted only weak influences on $F_{\text{WSO}}N$. In contrast, ALWC showed a decreasing trend across the three cities that was consistent with the regional variation of $F_{\text{WSO}}N$, and within each city $F_{\text{WSO}}N$ increased with increasing ALWC, indicating that ALWC plays a substantial role in modulating $F_{\text{WSO}}N$. We further emphasize that CD is located at the bottom of the SCB, where the unique deep-basin topography leads to stagnant winter conditions and frequent low boundary layers, resulting in continuous pollutant accumulation. Strong temperature inversions in CD promote increases in the low-level cloud liquid water path and near-surface RH, exerting a pronounced regulatory effect on ALWC accumulation (Guo et al., 2026). The coupling of these topographic and meteorological conditions makes wintertime ALWC in CD significantly higher than in other regions, making the regulatory effect of ALWC on WSON gas-particle partitioning more pronounced than in coastal or northern Chinese cities. All revisions have been marked in blue in the revised manuscript. We hope this adequately addresses your concern.

Revised text as it appears in the text (Page 11, line 278-297):

“Table 2 presents the differences in $F_{\text{WSO}}N$ across three Chinese cities, namely CD (representing the SCB), SH (representing the YRD), and Xianghe (XH, representing the NCP). As shown in the Table 2, $F_{\text{WSO}}N$ was highest in CD (0.70 ± 0.14), followed by SH (0.49 ± 0.26), and lowest in XH (0.43 ± 0.23). The variation in $F_{\text{WSO}}N$ among the three cities did not follow the same trends as T and RH, and no significant differences in pH were observed, suggesting that T, RH, and pH exerted only weak influences on $F_{\text{WSO}}N$. ALWC showed a decreasing trend across the three cities, which was consistent with the regional variation trend of $F_{\text{WSO}}N$. In addition, it

was also observed that within each city, $F_{\text{WSO}}N$ increased with increasing ALWC, implying that ALWC may play a substantial role in modulating $F_{\text{WSO}}N$ variations. As mentioned earlier, CD is located at the bottom of the SCB, and its unique deep-basin topography leads to unfavorable meteorological conditions in winter, such as stagnant conditions (Table 1) and the frequent occurrence of low boundary layers, resulting in the continuous accumulation of pollutants. The strong temperature inversions in CD promote increases in the low-level cloud liquid water path and near-surface RH, exerting a pronounced regulatory effect on the accumulation of ALWC (Guo et al., 2026). The coupling of these topographic and meteorological conditions makes the wintertime ALWC level in CD significantly higher than that in other regions, making the regulatory effect of ALWC on $F_{\text{WSO}}N$ gas-aerosol partitioning more pronounced than that in coastal or northern Chinese cities.

Table R1. Concentrations Regional comparison of key physicochemical parameters ($\text{PM}_{2.5}$, T, RH, pH, ALWC) and the water-soluble organic nitrogen partitioning coefficient ($F_{\text{WSO}}N$) in atmospheric aerosols at Chengdu (CD), Shanghai (SH), and Xianghe (XH).

Specie	City	Whole	Clean Period	Transition Period	Pollution Period
$F_{\text{WSO}}N$	CD	0.70 ± 0.14 (0.28–0.98)	0.62 ± 0.17 (0.29–0.88)	0.66 ± 0.14 (0.28–0.90)	0.73 ± 0.13 (0.42–0.98)
	SH	0.49 ± 0.26 (0.01–0.99)	0.37 ± 0.21 (0.01–0.93)	0.52 ± 0.29 (0.01–0.99)	0.64 ± 0.23 (0.15–0.97)
	XH	0.43 ± 0.23 (0.03–0.99)	0.33 ± 0.18 (0.03–0.69)	0.46 ± 0.20 (0.04–0.78)	0.48 ± 0.25 (0.05–0.99)
$\text{PM}_{2.5}$	CD	76 ± 38 (7–199)	29 ± 11 (7–49)	55 ± 18 (9–101)	100 ± 33 (30–199)
	SH	54 ± 40 (4–225)	23.9 ± 13.5 (4.0–91.0)	50 ± 26 (5–119)	100 ± 38 (35–225)
	XH	63 ± 56 (0–239)	18 ± 15 (0–69)	61 ± 27 (4–123)	118 ± 52 (1–239)
T	CD	6.4 ± 3.6 (-2.4–16.5)	6.2 ± 2.7 (0.2–12.3)	6.4 ± 3.4 (-1.3–15.4)	6.4 ± 3.9 (-2.4–16.5)
	SH	7.8 ± 3.9 (-1.2–19.9)	8.6 ± 4.4 (-1.2–19.9)	7.6 ± 3.4 (-0.91–16.6)	7.1 ± 3.8 (0.2–17.7)
	XH	-2.0 ± 5.2 (-14.2–9.8)	-5.3 ± 4.6 (-14.2–6.5)	-0.8 ± 3.9 (-7.6–9.8)	0.9 ± 4.3 (-8.3–9.1)
RH	CD	78 ± 19 (20–100)	78 ± 22 (26–99)	76 ± 19 (20–100)	79 ± 18 (29–100)
	SH	83 ± 14 (42–100)	88 ± 11 (53–100)	83 ± 14 (45–100)	77 ± 15 (42–98)
	XH	54 ± 21 (16–92)	47 ± 20 (24–91)	60 ± 21 (16–92)	57 ± 19 (18–92)
pH	CD	4.4 ± 0.2 (3.4–5.0)	4.2 ± 0.3 (3.4–4.5)	4.4 ± 0.2 (3.6–4.9)	4.4 ± 0.2 (3.8–5.0)
	SH	4.2 ± 0.3 (3.2–4.8)	4.0 ± 0.3 (3.3–4.8)	4.2 ± 0.3 (3.2–4.6)	4.4 ± 0.2 (3.8–4.8)
	XH	5.0 ± 0.9 (1.7–8.9)	4.5 ± 0.8 (1.7–6.0)	5.2 ± 0.6 (4.6–7.6)	5.4 ± 0.7 (3.8–8.9)
ALWC	CD	64.0 ± 71.5 (0.5–422.2)	34.0 ± 35.8 (1.2–130.5)	44.4 ± 49.2 (0.5–215.9)	87.6 ± 85.1 (1.6–422.2)
	SH	51.6 ± 51.4 (2.0–329.4)	23.2 ± 22.4 (2.0–130.1)	45.2 ± 37.6 (5.4–165.8)	80.4 ± 64.3 (6.6–329.4)
	XH	38.7 ± 73.6 (0.0–439.8)	9.4 ± 17.7 (0.0–103.3)	33.9 ± 36.2 (0.1–129.6)	73.2 ± 106.2 (0.0–439.8)

(Table R1 presents in the manuscript as Table 2)”

76 **Comments:**

77 My second concern is the amine partitioning. While the manuscript notes the
78 enhanced partitioning of methylamine (MA) and dimethylamine (DMA) into the
79 particle phase during polluted periods, it does not explore the underlying mechanisms
80 beyond physical partitioning. It remains unclear whether the observed MA/DMA
81 partitioning reflects purely physical dissolution or if it is linked to chemical
82 consumption via reactions in the aqueous phase. Distinguishing between physical
83 partitioning and chemical-enhanced partitioning would significantly elevate the
84 mechanistic insight of the paper. Furthermore, the discussion should address how
85 changes in aerosol pH (resulting from NH₃ mitigation) might non-linearly affect
86 amine partitioning and secondary formation via aqueous chemistry in this specific
87 high-humidity environment. This would bridge thermodynamic partitioning with
88 chemical transformation.

89 **Reply:** We sincerely thank you for this insightful comment. We agree that
90 distinguishing between physical partitioning and chemical-enhanced partitioning, and
91 discussing how changes in aerosol pH may non-linearly affect amine partitioning and
92 secondary formation, would significantly strengthen the mechanistic insight of this
93 study.

94 We clarify the distinction between physical partitioning and chemical-enhanced
95 partitioning. In our observations, MA and DMA were predominantly present in the
96 gas phase, but their partitioning into the particle phase was significantly enhanced
97 during polluted periods. This enhancement was closely associated with ALWC and
98 the presence of acidic inorganic species. In addition to physical dissolution into the
99 particle phase, aqueous-phase chemical reactions also enhance the partitioning
100 behavior. Gaseous amines dissolve into the aerosol liquid water following Henry's law,
101 and the dissolved amines then undergo rapid protonation within the aqueous phase to
102 form aminium ions, which subsequently react with sulfate and nitrate to form stable
103 aminium salts. This acid-base neutralization effectively converts volatile free bases
104 into non-volatile particulate aminium salts, thereby shifting the partitioning
105 equilibrium toward the particle phase.

We emphasize the role of ALWC in modulating amine partitioning. ALWC provides the aqueous medium necessary for both the dissolution and protonation of amines, and our observations show that the partitioning coefficients of MA and DMA increased significantly with increasing ALWC. The dominant contribution of NH_4NO_3 to ALWC, approximately 61%, further implies that nitrate-driven aerosol water indirectly enhances amine partitioning, establishing a positive feedback between inorganic aerosol formation and organic nitrogen partitioning in the SCB.

Regarding the potential non-linear effect of aerosol pH changes, resulting for example from NH_3 mitigation, on amine partitioning and secondary formation, we have carefully considered this point. In this study, the observed $\text{PM}_{2.5}$ pH range was 3.4 to 5.0. Within this range, amines are almost entirely protonated, and their partitioning coefficients showed no significant dependence on pH (Fig. R1). This is consistent with Tilgner et al. (2021), who stated that since atmospheric waters are typically acidic, bases are predominantly present in their protonated form, and their partitioning is not greatly altered by typical variations in pH. Therefore, under the specific high-humidity conditions of CD, pH changes within the observed range are unlikely to exert a strong non-linear control on amine partitioning. However, we acknowledge that if NH_3 mitigation were to shift aerosol pH into a less acidic regime, the protonated fraction and thus the partitioning behavior could change non-linearly, and future studies with a wider pH range are therefore needed to fully resolve this issue.

All revisions have been marked in blue in the revised manuscript. We hope this adequately addresses your concern.

Revised text as it appears in the text (Page 12-13, line 312-357):

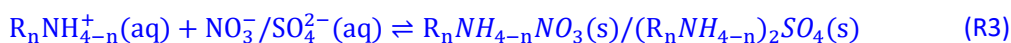
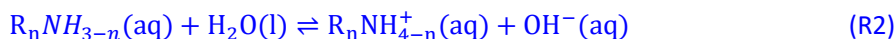
“Amines contribute significantly to $\text{WSO}_{\text{g,p}}$ in CD. Specifically, amines-N (MA-N and DMA-N) account for 48.7% and 10.5% of WSON in the gas and particle phases, respectively (Fig. 4). During the winter haze periods in CD, the total concentration of amines increased substantially, and the gas-particle partitioning coefficient (F) nearly doubled (Table 1), indicating enhanced gas-to-particle

conversion. It is also evident from Table 1 that the gas-aerosol partitioning coefficients of the basic nitrogen-containing species (NH_4^+ , MA, DMA) followed a decreasing order of F_{AM} (0.36) > F_{MA} (0.25) > F_{DMA} (0.18), indicating that as the hydrogen atoms on the amino group ($-\text{NH}_2$) are progressively substituted by hydrophobic methyl groups ($-\text{CH}_3$), these basic species increasingly tend to remain in the gas phase rather than partitioning into the aerosol phase. Previous studies have demonstrated that with increasing degree of methylation of amines, their relative hydration free energy increases, thereby weakening their affinity for water and enhancing their partitioning from the aerosol phase to the gas phase (Rizzo and Jorgensen, 1999). In addition, as shown in Table S2, the reaction rate of NH_3 with hydroxyl radical ($\cdot\text{OH}$) in the atmosphere is relatively slow ($1.6 \times 10^{-13} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$), corresponding to an atmospheric lifetime of up to 72.3 days. In contrast, MA ($(2.20 \pm 0.22) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) and DMA ($(6.54 \pm 0.66) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) react extremely rapidly with $\cdot\text{OH}$, with lifetimes of only a few hours (12.6 h and 4.3 h for MA and DMA, respectively) (Table S2). This implies that the fast chemical loss of their gas-phase concentrations directly reduces their accumulation potential in the aerosol phase, leading to the observed decline in the measured F values with increasing methylation degree.

To investigate the partitioning behavior of amines, we calculated their partitioning coefficients under different diel conditions, as shown in Fig. 5. As depicted in Figs. 5a and 5c, OM exhibited no significant correlation with the F_p of amines during either daytime or nighttime ($R < 0.5$, $p > 0.05$), indicating that OM exerted only a weak influence on the gas-aerosol partitioning of MA and DMA, and that their partitioning into the aerosol organic phase was relatively low. In contrast, both F_{MA} and F_{DMA} showed a clear increasing trend with increasing ALWC ($R > 0.5$, $p < 0.05$), suggesting that MA and DMA preferentially partitioned into the aerosol aqueous phase.

Furthermore, we found significant linear relationships between F_p and acidic anions (nitrate and sulfate) ($R > 0.5$, $p < 0.01$), and the coefficients increased with NOR and SOR (Fig. 6). This indicates that inorganic salts significantly promote the

partitioning of MA and DMA, and that reactive uptake may also serve as one of the driving forces facilitating the partitioning of amines into aerosol. According to the Reactions (9 – 12) (Ge et al., 2011a), gaseous amines synergistically neutralize H₂SO₄/HNO₃ together with NH₃ to generate particulate amine salts (Almeida et al., 2013; Glasoe et al., 2015; Liu et al., 2018). Figure S5 presents a comparison between amines and inorganic salts. Our results show that amines exhibited good linear correlations with both nitrate and sulfate ($R > 0.5$, $p < 0.01$), suggesting that MA and DMA are likely present in particles predominantly as amine nitrate and/or amine sulfate salts (Huang et al., 2022). Furthermore, ALWC was found to promote their formation (Fig. S5). The humid and stagnant meteorological conditions in CD (Table 1) provide an ideal environment for the above transformations, promoting efficient enrichment of amines in the aerosol phase and making them a significant component of water-soluble ions in PM_{2.5}.



(Eq. R1-R4 presents in the manuscript as Eq. 9-12)”

Table R2. Atmospheric physicochemical properties of different alkaline nitrogen-containing compounds (25 °C)

	Ammonia	Methylamine	Dimethylamine
CAS	7664-41-7	74-89-5	124-40-3
Molecular weight	17	31	45
pKa (Huang, 2022)	9.25	10.66	10.73
Vapor pressure (kPa)	1003 (Youn et al., 2015)	350.6 (He et al., 2026)	203 (Youn et al., 2015)
Henry's law constant (mol atm ⁻¹ kg ⁻¹) (Sander, 2023)	59.96	90.179	56.742
Reaction rate of ·OH (cm ³ molecule ⁻¹ s ⁻¹)	1.6×10 ⁻¹³	(2.20±0.22)×10 ⁻¹¹	(6.54±0.66)×10 ⁻¹¹
[half-life] (Ge et al., 2011b)	[72.3 d]	[12.6 h]	[4.3 h]

(Table R2 presents in the manuscript as Table S2)

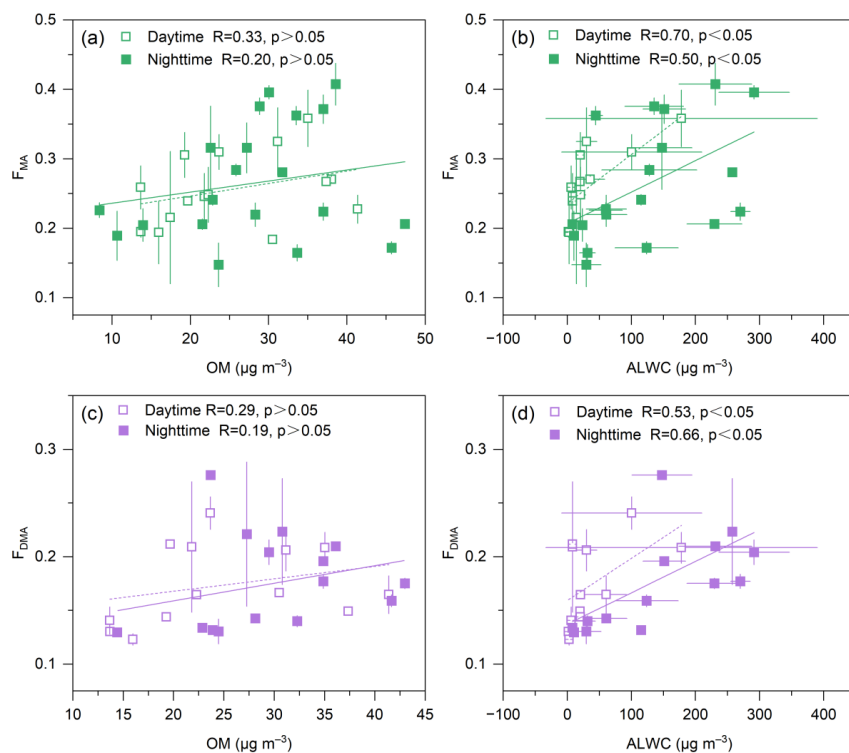


Figure R1. Linear regression analysis for F_p with OM and ALWC under the daytime and nighttime during the pollution period (2025.1.2—2025.1.6). (Figure R1 presents in the manuscript as Figure 5)

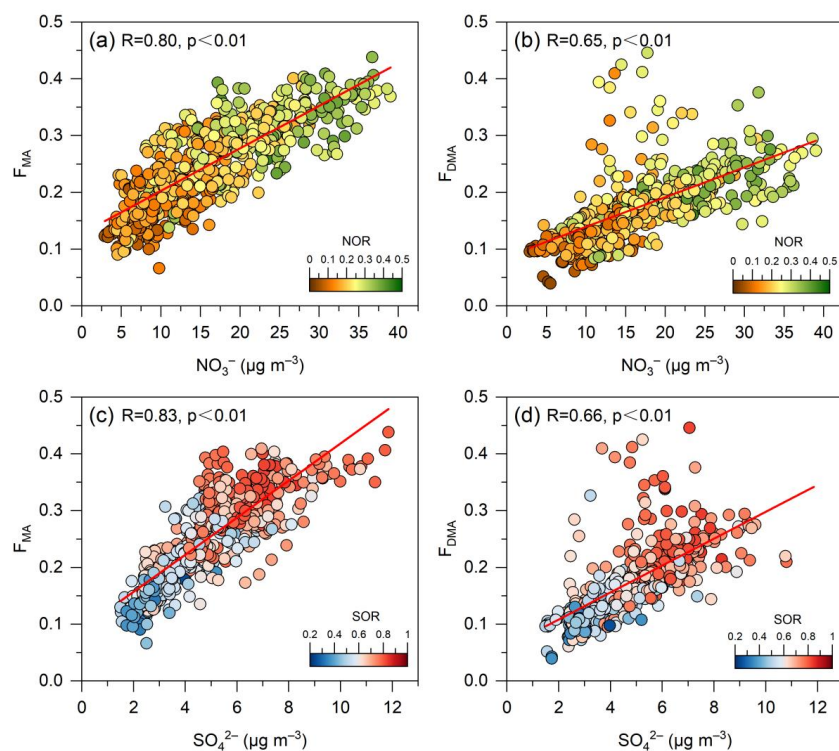
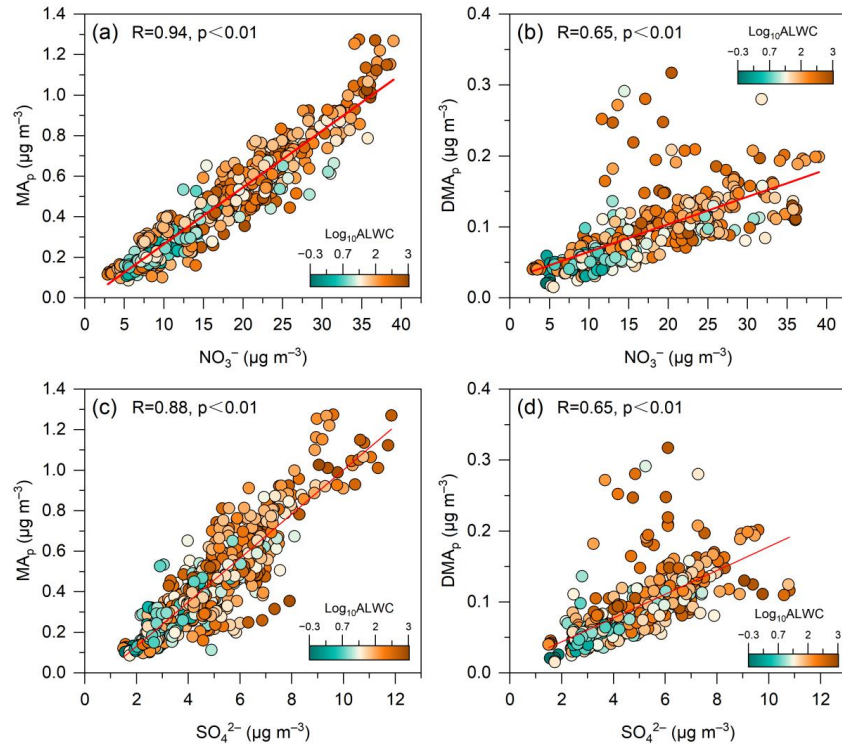


Figure R2. Linear regression analysis for the gas-aerosol partitioning coefficient (F_p)

188 of amines with inorganic salts. (Figure R2 presents in the manuscript as Figure 6)



189

190 **Figure R3.** Linear regression analysis for the amines with inorganic salts. (Figure R2
191 presents in the manuscript as Figure S5)

192 **Comments:**

193 Another concern is the partitioning coefficient (F_p) dynamics. While the manuscript's
194 most notable contribution is the provision of field-derived $F_p(\text{WSN})$ data, I am
195 concerned that the subsequent analysis with drivers of the partitioning coefficient
196 itself. Throughout the Results and Discussion, the authors attribute variations in
197 particulate WSN (C_p) and gaseous WSN (C_g) to factors influencing F_p . However,
198 by using C_p (or C_g) as the primary dependent variable in correlation analyses and
199 S-curve fittings, the manuscript conflates concentration dynamics with partitioning
200 thermodynamics. To rigorously isolate F_p drivers, the authors should redefine the
201 dependent variable as F_p . Without this distinction, the claim that ALWC “drives” F_p
202 remains circumstantial.

203 **Reply:** We sincerely thank you for this important concern. We agree that using C_p or
204 C_g as the primary dependent variable in correlation analyses and S-curve fittings can

conflate concentration dynamics with partitioning thermodynamics, and that F_p should be defined as the dependent variable when isolating the drivers of partitioning. Therefore, in the revised manuscript, we have deleted the relevant statements on the dynamics of WSON and the partitioning coefficient F_p in light of your subsequent suggestions. The revised manuscript no longer attributes variations in C_p or C_g to factors influencing F_p , and it no longer uses C_p or C_g as the dependent variable to infer the drivers of F_p . Any remaining discussion of partitioning drivers is now based directly on F_p as the dependent variable. For the S-curve analysis of amines, the dependent variable is the theoretical partitioning coefficient itself, rather than C_p or C_g . For ALWC, we have now added a scatter plot to directly discuss the relationship between ALWC and F_p (Fig. R1), and we have adjusted the wording of the conclusions accordingly to avoid overstating causality. All revisions have been marked in blue in the revised manuscript. We hope this adequately addresses your concern.

Revised text as it appears in the text (Page 12-13, line 334-341):

“To investigate the partitioning behavior of amines, we calculated their partitioning coefficients under different diel conditions, as shown in Fig. 5. As depicted in Figs. 5a and 5c, OM exhibited no significant correlation with the F_p of amines during either daytime or nighttime ($R < 0.5$, $p > 0.05$), indicating that OM exerted only a weak influence on the gas-aerosol partitioning of MA and DMA, and that their partitioning into the aerosol organic phase was relatively low. In contrast, both F_{MA} and F_{DMA} showed a clear increasing trend with increasing ALWC ($R > 0.5$, $p < 0.05$), suggesting that MA and DMA preferentially partitioned into the aerosol aqueous phase.”

Specific Comments:

Comments:

Lines 75-77: The authors state here that the gas-particle partitioning of amines is governed by multiple factors, including aerosol pH, relative humidity, and the

composition of the condensed phase. This statement is of interest and warrants further discussion / interpretation. Since the gas- and particle-phase amines have been measured in this manuscript, I would recommend the authors providing an in-depth discussion on their partitioning mechanism in the text.

Reply: We thank you for this valuable suggestion. We agree that, since both gas- and particle-phase amines were measured in this study, an in-depth discussion of their gas-particle partitioning mechanism is necessary and meaningful. All revisions have been marked in blue in the revised manuscript. We sincerely appreciate your careful review and constructive suggestion.

Comments:

Introduction: On the whole, I believe that this section could be greatly improved. It could be more focused to better serve the specific aims of this study. And the recent findings on the WSON or WSOC gas-to-particle partitioning observation, particularly the authors' own work should also be summarized.

Reply: We sincerely thank you for this constructive suggestion. We agree that the Introduction should be more focused and better aligned with the specific aims of this study, and that recent observational findings on WSON and WSOC gas-to-particle partitioning, particularly our own previous work, should also be summarized. In the revised manuscript, we have substantially restructured and condensed the Introduction by removing overly broad background material and reorganizing the text so that it moves more directly from the atmospheric importance of organic nitrogen and WSON to the central scientific issue of this study, namely the gas-particle partitioning of WSON and amines. We now explicitly state that most previous studies have measured only particle-phase WSON, whereas simultaneous gas- and particle-phase observations remain scarce and the factors controlling WSON and amine partitioning are still not well constrained. We have also added a concise summary of observational studies on WSOC gas-to-particle partitioning, particularly our own previous work (e.g., Lv et al., 2022a, b; Li et al., 2025), which places the present study in the context of existing observational evidence and highlights its novelty. Finally, the Introduction

now ends with a focused statement of the specific aims of this study: to simultaneously measure gas- and particle-phase WSON and amines in the Sichuan Basin, to quantify their partitioning coefficients, and to elucidate the key drivers, especially ALWC, pH, and temperature, under winter conditions. All revisions have been marked in blue in the revised manuscript, and we hope these changes adequately address your concern.

Revised text as it appears in the text (Page 3-4, line 39-94):

“Organic nitrogen (ON) is a ubiquitous component of fine particulate matter (PM_{2.5}), accounting for 10%–50% of total nitrogen (TN) in global aerosols (Leung et al., 2024). Atmospheric nitrogen-containing organic compounds (NOCs) encompass a wide range of species, including amines, N-heterocycles, nitroaromatics, organic nitrates, and so on (Jickells et al., 2013; Yu et al., 2024). These species partition between the gas and condensed phases depending on their volatility and water solubility. For example, low-molecular-weight amines and nitroaromatic compounds are semi-volatile and partition dynamically between phases (Liu et al., 2018; Chen et al., 2025), while monofunctional organic nitrates predominantly reside in the gas phase, whereas multifunctional ones preferentially partition into the particle phase (Rollins et al., 2013; González-Sánchez et al., 2021). Among these, water-soluble organic nitrogen (WSON) is of particular interest, as it actively participates in aqueous-phase reactions and significantly contributes to secondary organic aerosol (SOA) formation (Yu et al., 2020).

Amines, as a major class of water-soluble NOCs in the atmosphere, play a pivotal role in aerosol formation and evolution. The H₂SO₄-amine-H₂O ternary nucleation mechanism markedly enhances new particle formation (NPF) rates by stabilizing critical clusters, with ambient observations demonstrating more than a thousand-fold increase in formation rates compared to ammonia (Almeida et al., 2013; Schobesberger et al., 2013; Yao et al., 2018; Kanawade and Jokinen, 2025; Yu et al., 2025). Through acid-base reactions, amines readily form stable aminium salts with inorganic acids, altering particles hygroscopicity, density, and optical properties (Qiu

and Zhang, 2013). Additionally, Maillard-like reactions between amines and carbonyls in the aqueous phase contribute substantially to brown carbon (BrC) formation (Tang et al., 2022; Xiao et al., 2025). Despite these recognized roles, the gas-aerosol partitioning behavior of amines remains critically underexplored. Previous studies suggest that amine partitioning is influenced by aerosol acidity (pH), relative humidity (RH), and condensed-phase composition, and is strongly pH-dependent, favoring the particle phase under acidic conditions (Ge et al., 2011a; Pankow, 2015). However, amine gas-aerosol partitioning remains poorly understood, hindering accurate assessments of their contributions to PM formation and climate, and thus warranting urgent investigation.

In contrast, the gas-aerosol partitioning of bulk water-soluble organic compounds (WSOC) has been more extensively investigated. Recent simultaneous online measurements in the Yangtze River Delta (YRD) and North China Plain (NCP) have systematically elucidated the driving mechanisms of WSOC partitioning (Lv et al., 2022a; Lv et al., 2022b; Li et al., 2025; Li et al., 2026). These studies revealed that under dry conditions ($RH < 80\%$), WSOC partitioning is governed by temperature (T) and proceeds via condensation into the organic phase; under humid conditions ($RH > 80\%$), it shifts to being dominated by aerosol liquid water content (ALWC), with uptake into the aqueous phase (Lv et al., 2022a). Ammonia (NH_3) not only elevates particle pH through neutralization to promote organic acid dissociation, but also participates in aqueous-phase reactions to form nitrogen-containing organics (e.g., imidazoles), contributing to WSON and BrC light absorption (Lv et al., 2022a; Li et al., 2026). Nationwide data show a strong linear correlation ($R = 0.77$) between $WSOC_p$ and NH_4NO_3 mass fractions, confirming the critical role of NH_3 in regional SOA formation (Lv et al., 2022a). However, the gas-aerosol partitioning behavior of WSON has received far less attention, with most studies limited to particle-phase measurements and thus unable to capture the dynamic exchange between phases.

This knowledge gap is especially pronounced in regions with complex aerosol chemistry and high precursor emissions, such as the Sichuan Basin (SCB), a topographically enclosed basin characterized by high RH, frequent winter haze, and

substantial agricultural and industrial NH_3 emissions. To address this gap, we conducted an intensive winter observation campaign in Chengdu (CD) from December 2024 to January 2025, simultaneously measuring gas- and particle-phase WSON and two amines. Our work aims to elucidate the pollution characteristics of WSON and amines, and the driving factors governing their gas-particle partitioning behavior in the atmosphere.”

Comments:

Materials and methods: The manuscript currently describes the sampling site only in terms of its urban location, coordinates, and surrounding land use, and lacks meteorological context relevant to partitioning. Given the central variable of this study (i.e., $F_p(\text{WSON})$) is highly sensitive to relative humidity, temperature, boundary layer height and ALWC and chemical composition of aerosol, all of which are strongly shaped by the topographic confinement of the Sichuan Basin. So I suggest the authors add details on basin meteorology and the unique emission characteristics of Chengdu to justify the site’s selection.

Reply: We sincerely thank you for this valuable suggestion. We agree that the Materials and Methods section should provide the meteorological and emission context needed to justify the site selection, given that F_p (WSON) is highly sensitive to relative humidity, temperature, boundary layer height, ALWC, and aerosol chemical composition, all of which are strongly shaped by the topographic confinement of the Sichuan Basin. In the revised manuscript, we have added a detailed description at the end of **Section 2.1 Observation site description**. Specifically, we now describe the deep-basin topography of the Sichuan Basin, which severely hinders pollutant dispersion, and the winter meteorological conditions, including high relative humidity, frequent fog, persistent temperature inversions (winter inversion frequency of 95.4% and depth of 289.4 m), and a boundary layer height that can drop below 500 m with relative humidity frequently exceeding 70%, thereby promoting hygroscopic growth and secondary transformation. We also summarize the unique emission characteristics of Chengdu, where $\text{PM}_{2.5}$ sources are

dominated by vehicular emissions (25.6%) and secondary sources (40.5%), with secondary inorganic ions accounting for 40% to 60% of PM_{2.5} mass in winter and approximately half of that being nitrate, which provides abundant precursors for the secondary formation of nitrogen-containing organics. We further explain that these coupled topographical, meteorological, and emission characteristics endow Chengdu with unique regional representativeness for investigating the gas-particle partitioning of nitrogen-containing organic compounds under stagnant winter conditions, and that our observation site in the central urban area at 25 m above ground can effectively capture the influence of these basin-specific processes on WSON partitioning. All revisions have been marked in blue in the revised manuscript. We appreciate your careful review and hope these additions adequately address your concern.

Revised text as it appears in the text (Page 5, line 104-125):

“The SCB features a typical deep-basin topography (with a basin depth exceeding 2000 m), surrounded by mountains on all sides, which severely hampers the dispersion of air pollutants (Rao et al., 2026). In winter, the Qinling Mountains block the intrusion of cold air, allowing warm and moist air masses to accumulate within the basin, leading to high humidity, frequent fog, and recurrent temperature inversions (with a winter inversion frequency of 95.4% and an inversion layer depth of 289.4 m) (Feng et al., 2020). During pollution episodes, the boundary layer is significantly compressed, with its height reduced by approximately 50% (Lu et al., 2024), resulting in a stable atmospheric stratification that is extremely unfavorable for pollutant diffusion. The synergistic effects of increased emissions, stagnant meteorological conditions, and enhanced aqueous-phase reactions under high relative humidity (> 80%) can readily cause extreme PM_{2.5} episodes (Shu et al., 2025), and the humid environment further promotes the hygroscopic growth and secondary transformation of particulate matter (Wang et al., 2021). In CD, the major sources of PM_{2.5} are vehicle emissions (25.6%) and secondary sources (40.5%) (Zhang et al., 2024). In winter, secondary inorganic ions account for 40% – 60% of PM_{2.5} mass, of which approximately half is nitrate (Rao et al., 2026), providing abundant precursors

for the secondary formation of NOCs (Li et al., 2023). The combined topographic-meteorological-emission characteristics impart unique regional representativeness to the study of gas-aerosol partitioning of NOCs under the humid and stagnant conditions typical of CD. Therefore, the observation site can effectively capture the influence of the basin-specific atmospheric chemical processes on the partitioning of WSON.”

Comments:

Lines 110-134: While the OC-to-OM conversion factor ($k=1.6$) is mentioned in Table 1, the Methods section contains no explicit statement on how OM was derived from measured OC. F_p definition should also appear in this section, not only in Results/Figure captions.

Reply: We sincerely thank you for pointing out these omissions. We agree that the Methods section should explicitly state how OM was derived from the measured OC and should also provide the definition of F_p rather than leaving it only in the Results and figure captions. In the revised manuscript, we have added a clear statement in the Methods section that the organic matter concentration was calculated from the measured organic carbon using $OM = OC \times 1.6$, and we have also included the definition of the gas-particle partitioning coefficient as $F_p = C_p / (C_g + C_p)$, where C_p and C_g are the concentrations of the target species in the particle phase and gas phase, respectively. All revisions have been marked in blue in the revised manuscript. We appreciate your careful review and hope these additions adequately address your concern.

Revised text as it appears in the text (Page 6, line 142-143):

“The gas-particle phase partition coefficient (F_p) for WSON is calculated as follows, similarly, similar calculations apply to amines:

$$F_p = \frac{C_p}{C_g + C_p} \quad (R5)$$

(Eq. R5 presents in the manuscript as Eq. 4)”

Revised text as it appears in the text (Page 7, line 174-175):

“The contribution of organic compounds to ALWC (ALWC_{org}) was determined using the mass concentration of organic matter (OM) which was calculated on the basis of OC ($OM = OC \times 1.6$) (Lv et al., 2022a; Lv et al., 2022b), as described in detail in the Supporting Information Text S2.”

Comments:

Lines 197-241: The inference that “more NOCs are formed during the accumulation of OM” from an increasing WSON/WSOC_p ratio is invalid. Since WSOC accumulates substantially via secondary formation of non-nitrogenous organics during haze, this ratio merely signals the relative enrichment of nitrogenous species, not absolute NOC formation. Additionally, attributing higher daytime WSON to photochemistry lacks supporting precursor/radical data. Dismissing the clean-day trend as a “lack of photochemistry” is inconsistent, as radicals persist on clean days; diurnal variations are more plausibly explained by boundary layer dynamics and emission patterns.

Reply: We sincerely thank you for this careful and insightful comment. We fully agree that inferring enhanced absolute NOC formation from an increasing WSON/WSOC_p ratio is not valid, because WSOC can accumulate substantially through secondary formation of non-nitrogenous organics during haze episodes, so this ratio only indicates the relative enrichment of nitrogenous species rather than the absolute formation of NOCs. We also agree that attributing higher daytime WSON to photochemistry lacks support from precursor or radical measurements, and that dismissing the clean-day trend as a lack of photochemistry is inconsistent, since radicals persist on clean days and diurnal variations are more plausibly explained by boundary layer dynamics and emission patterns. In the revised manuscript, we have changed the relevant statement to “Notably, the WSON/WSOC_p ratio increases as pollution worsens (Fig. S3i–l), indicating that more NOCs are enriched during the accumulation of OM.” We have also deleted the entire discussion concerning the diurnal variation of WSON and its attribution to photochemistry. All revisions have

been marked in blue in the revised manuscript. We appreciate your rigorous review and hope these changes adequately address your concern.

Revised text as it appears in the text (Page 9, line 220-222):
“Notably, the WSON/WSOC_p ratio increases as pollution worsens (Fig. S3i–l), indicating that more NOCs are enriched during the accumulation of OM.”

Comments:

Lines 242-264: The assertion that photochemistry rapidly generates WSON_g during the daytime is not supported by precursor or radical data. The ALWC during the daytime may also influence the partitioning of WSON. Furthermore, the claim of “no significant correlations ($R < 0.3$)” between F_p and T, RH, or pH fails to specify whether Pearson (linear) or Spearman (monotonic) coefficients were used. Explaining the modest ALWC– F_p correlation in Chengdu via an “organic shell hindering gas transfer” is speculative.

Reply: We sincerely thank you for these careful and constructive comments. We agree that the assertion that photochemistry rapidly generates WSON_g during the daytime is not supported by precursor or radical data, and that ALWC during the daytime may also influence the partitioning of WSON. In the revised manuscript, we have therefore deleted the entire discussion of the diurnal variation of WSON and its attribution to photochemistry. Regarding the correlation analysis, we confirm that the original analysis employed the Pearson linear correlation coefficient. To better examine both linear and nonlinear relationships between F_p and T, RH, and pH, we have replaced the original correlation heatmap with scatter plots (Fig. R4). For methylamine, the partitioning coefficient shows no significant linear or nonlinear dependence during either daytime or nighttime, and the same is true for dimethylamine during daytime. For dimethylamine at night, the partitioning coefficient increases exponentially with increasing RH, whereas the influences of T and pH are relatively weak. Finally, we agree that explaining the modest correlation between ALWC and F_p in Chengdu via an organic shell hindering gas transfer is speculative, and we have removed this

statement. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope these changes adequately address your concerns.

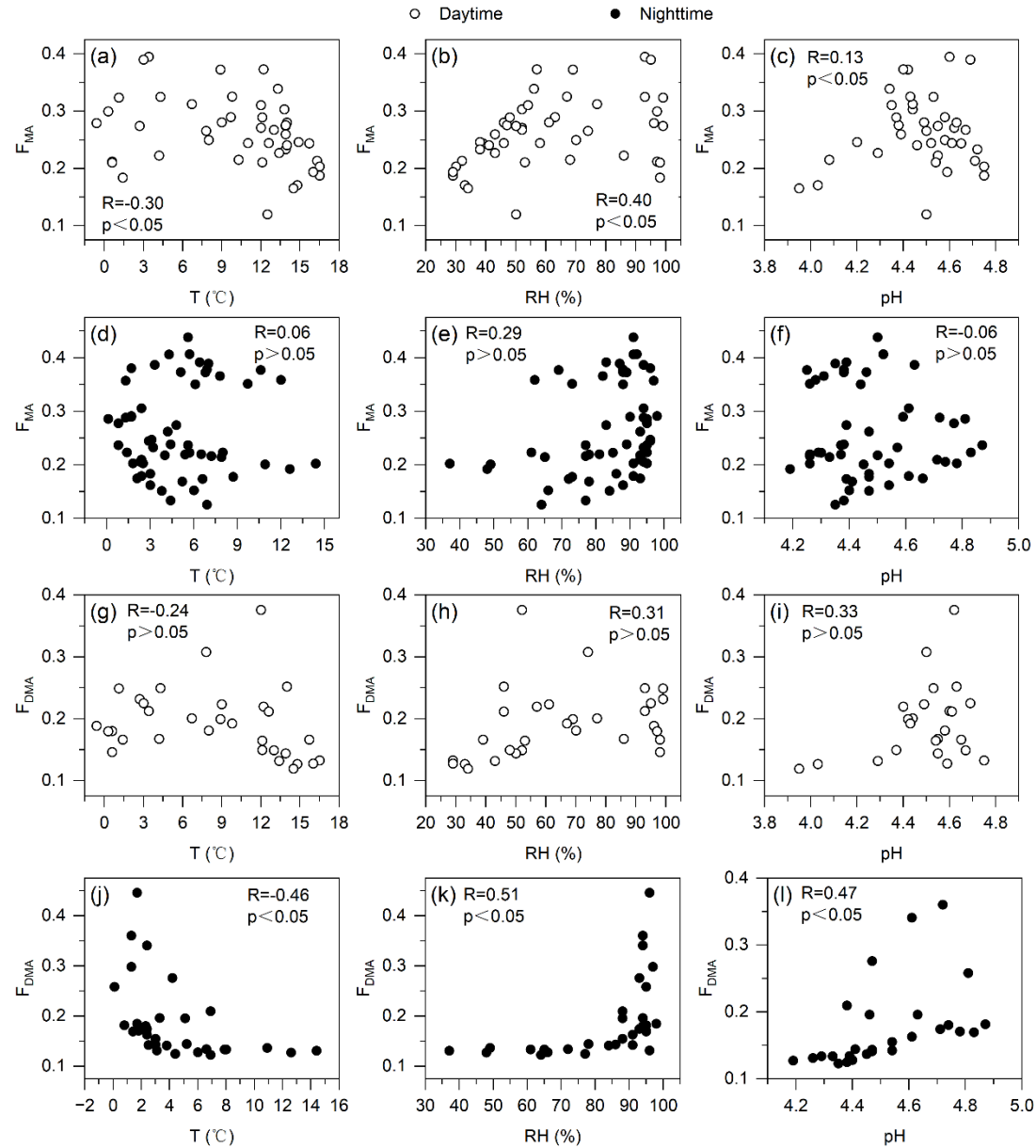


Figure R4. Factors affecting the gas-to-aerosol phase-partitioning coefficients of methylamine (F_{MA}) and dimethylamine (F_{DMA}) in Chengdu (CD) under the daytime and nighttime during the pollution period (2025.1.2—2025.1.6). (Figure R4 presents in the manuscript as Figure S6)

Comments:

Lines 319-355: Again, the correlation analysis fails to specify the type of correlation

coefficient used and address non-linear relationships, particularly for S-curve interpretations.

Reply: We thank you for this comment. We confirm that the original correlation analysis employed the Pearson linear correlation coefficient. To address non-linear relationships, particularly for the S-curve interpretations, we have replaced the original correlation heatmap with scatter plots. This is the same figure used to address your previous comment. For methylamine, the partitioning coefficient shows no significant linear or nonlinear dependence during either daytime or nighttime, and the same is true for dimethylamine during daytime. For dimethylamine at night, the partitioning coefficient increases exponentially with increasing RH, whereas the influences of T and pH are relatively weak. Regarding the S-curve interpretation, we have revised our reading of Fig. R1 and Fig. R4 (Figure 5 and Figure S6 in the manuscript, respectively). The observed pH range of 3.4 to 5.0 of PM_{2.5} falls within the flat plateau region of the sigmoidal curves, where the protonated fraction is approximately 100 percent. Therefore, within the observed pH range and the uncertainty of the available measurements, no significant independent pH dependence was detected. All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

Revised text as it appears in the text (Page 13, line 358-366):

“To further identify the key factors governing the partitioning of amines, we analyzed the correlations between F_p and meteorological conditions (i.e., RH and T) as well as pH. As shown in Figures S6a–f, no significant linear or nonlinear correlations were observed between F_{MA} and T, RH, or pH during either daytime or nighttime ($R < 0.5$). Similarly, F_{DMA} did not show notable correlations with T, RH, or pH during the daytime ($R < 0.5$, $p > 0.05$). However, during the nighttime, F_{DMA} exhibited a significant nonlinear relationship with RH, with an exponential increase when RH exceeded 80%, while the influences of T and pH remained relatively weak ($R < 0.5$, $p > 0.05$).”

Comments:

Section 3.3: The observation that MA and DMA shift to the particle phase during polluted periods is presented without mechanistic depth. The authors discuss ALWC, pH, and T as factors for MA/DMA partitioning, but treats amines as a bulk class and omits species-specific controls: protonation equilibrium, effective Henry's law constants, dicarbonyl-driven consumptive partitioning to imidazoles, organic-phase partitioning under high OM/inorganic ratio, aerosol aging time, and ionic-strength effects on activity coefficients. Without these, the conclusion that "ALWC is the key factor" remains a single-variable approximation of a multi-parameter thermodynamic problem.

Reply: We sincerely thank you for this insightful comment. In the revised manuscript, we have substantially expanded the mechanistic discussion in **Section 3.3** to address your concerns.

In the revised manuscript, we treat MA and DMA as distinct species rather than as a bulk amine class. Table R2 compares key physicochemical parameters of ammonia, MA, and DMA, including pKa, vapor pressure, Henry's law constant, and reaction rate with $\bullet$ OH. The pKa increases gradually, indicating increasing basicity and a stronger ability to accept protons, and thus a higher degree of protonation after entering the particle phase. Meanwhile, with increasing degree of methylation, the vapor pressure decreases, hydrophobicity increases, the reaction rate with OH radicals increases, and the atmospheric lifetime shortens, ultimately leading to the gas-particle partitioning coefficient order of $F_{AM} (0.36) > F_{MA} (0.25) > F_{DMA} (0.18)$. The partitioning behavior of amines in the atmosphere is shown in Fig. R5. Once gaseous amines enter the particle phase, they are rapidly protonated to form $R_n-NH_{4-n}^+$, and subsequently form stable clusters with acidic species such as inorganic salts (sulfate and nitrate) (Schobesberger et al., 2013; Yao et al., 2018). Free unprotonated amines ($R_n-NH_{3-n}(aq)$) are almost absent in the particle phase, so Henry's law plays only a minor role in controlling the partitioning between $R_n-NH_{3-n}(g)$ and $R_n-NH_{3-n}(aq)$. Tilgner et al. (2021) also stated in their original work: **"Since atmospheric waters are typically acidic, bases are predominantly present in their protonated form,**

and their partitioning is not greatly altered by typical variations in pH.” Fig. R1 illustrates the partitioning mechanism of amines in aerosols. It can be seen that F_p shows no significant linear or nonlinear relationship with OM, but exhibits a significant linear relationship with ALWC, increasing with increasing ALWC. In addition, Figs. R2, R3, and Reactions R1–R4 further indicate that reactive chemical uptake also enhances amine partitioning, and this enhancement increases with increasing NOR and SOR.

All revisions have been marked in blue in the revised manuscript. We appreciate your rigorous review and hope this adequately addresses your concern.

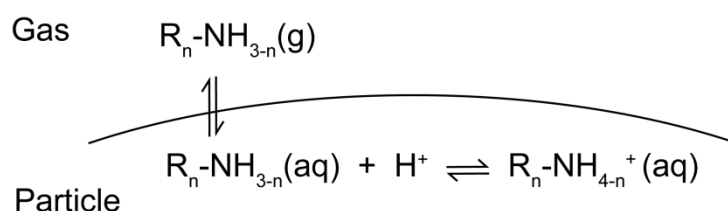


Figure R5. Schematic diagram of gas-particle distribution and electro-dissociation of amines.

Technical corrections:

Comments:

Author list appears to list Rui Li twice (positions 3 and 9), please confirm whether it is a metadata duplication.

Reply: Thank you for pointing this out. We confirm that this is not a metadata duplication. The two “Rui Li” entries correspond to two different individuals. The author listed in position 3 is Rui Li (李锐), a student at the School of Geographic Sciences, East China Normal University. The author listed in position 9 is Rui Li (李睿), a research professor at the School of Geographic Sciences, East China Normal University, who also holds a position at the Institute of Eco-Chongming, 20 Cuiniao Rd., Chongming, Shanghai, China. They share the same pinyin name but have different Chinese characters and are completely distinct persons.

Comments:

The unit of N should be consistently formatted as “ $\mu\text{gN m}^{-3}$ ” throughout the manuscript.

Reply: We thank you for pointing this out. We have carefully checked the entire manuscript and corrected the two instances where N was missing from the unit. The unit of N has now been consistently formatted as “ $\mu\text{gN m}^{-3}$ ” throughout the manuscript, including the abstract, main text, tables, figure captions, and Supplementary Information. All corrections have been marked in blue in the revised manuscript. We appreciate your careful review.

Revised text as it appears in the text (Page 6, line 139-140):

“The unit is $\mu\text{gN m}^{-3}$.”

Revised text as it appears in the text (Page 9, line 215-218):

“The concentration of $\text{WSO}\text{N}_\text{p}$ ($2.3 \pm 1.4 \mu\text{gN m}^{-3}$) was more than twice that of $\text{WSO}\text{N}_\text{g}$ ($1.2 \pm 0.9 \mu\text{gN m}^{-3}$) (Fig. 1c and Table 1), indicating that WSON in CD mainly exists in the particle phase, as observed at Mt. Fuji in Japan (Matsumoto et al., 2020).”

Comments:

Abbreviations (e.g., WSON, SOA, MA, DMA, ALWC) should be preceded by full names at their first appearance in the main text not only in the Abstract.

Reply: We thank you for pointing this out. We have carefully checked the entire manuscript and revised it accordingly. Full names are now provided for all abbreviations, including WSON, SOA, MA, DMA, and ALWC, at their first appearance in the main text, not only in the Abstract. In addition, we have compiled a summary table (Table R3) listing each abbreviation, its corresponding full name, and the line number of its first occurrence in the main text for your convenience. All revisions have been marked in blue in the revised manuscript. We appreciate your careful review.

Table R3. Abbreviations, full names, and line numbers of first occurrence in the revised manuscript.

No.	Abbreviation	Full name	Line number
1	ON	organic nitrogen	44
2	PM _{2.5}	particulate matter	44
3	TN	total nitrogen	45
4	NOCs	nitrogen-containing organic compounds	46
5	WSO _N	water-soluble organic nitrogen	54
6	SOA	secondary organic aerosol	56
7	NPF	new particle formation	59
8	BrC	brown carbon	66
9	pH	aerosol acidity	69
10	RH	relative humidity	69
11	WSOC	water-soluble organic compounds	74
12	the YRD	the Yangtze River Delta	76
13	the NCP	the North China Plain	76
14	T	temperature	79
15	ALWC	aerosol liquid water content	81
16	NH ₃	ammonia	82
17	the SCB	the Sichuan Basin	92
18	CD	Chengdu	95
19	MA	methylamine	132
20	DMA	dimethylamine	133
21	IGAC	In-situ Gas and Aerosol Compositions monitor	135
22	F _p	gas-particle phase partition coefficient	147
23	PP	pollution periods	154
24	CP	clean periods	155
25	TP	transition periods	155
26	OC	organic carbon	156
27	EC	elemental carbon	156
28	OM	organic matter	179
29	NOR	nitrogen oxidation ratio	195
30	SOR	sulfur oxidation ratio	195
31	SH	Shanghai	225
32	SNA	sulfate, nitrate, ammonium	256
33	XH	Xianghe	279
33	·OH	hydroxyl radical	326

Comments:

To avoid ambiguity, it is recommended to rephrase this expression “WSO_N transfer process” to “WSO_N partitioning process”.

Reply: We thank you for this suggestion. We agree that “WSO_N partitioning process” is clearer and avoids potential ambiguity. We have revised that instance accordingly,

and the change has been marked in blue in the revised manuscript. We appreciate your careful review.

Revised text as it appears in the text (Page 2, line 18):

“However, the factors influencing this WSON partitioning process remain unclear.”

Comments:

“the decrease in F_{WSON} during the daytime may be attributed to the increase of $WSON_g$ exceeding that of $WSON_p$ ” merely restates the mathematical definition. Please delete it or replace it with a mechanistic explanation.

Reply: We thank you for this comment. We agree that this sentence merely restates the mathematical definition of F_p . We have deleted it from the revised manuscript. We appreciate your careful review.

Comments:

The manuscript uses three different designations for the sampling location: “Chengdu”, “CD”, and “SCB”. The authors should standardize the nomenclature throughout the text, tables, and figures.

Reply: We thank you for pointing out the inconsistent nomenclature. We have standardized the designations throughout the manuscript. “Chengdu” is now used at its first appearance, and “CD” is used thereafter when referring to the sampling city and site. “SCB” is used exclusively when referring to the Sichuan Basin as a geographical region and is no longer used as a designation for the sampling location. We have checked and unified these terms in the main text, tables, figures, and figure captions. All revisions have been marked in blue in the revised manuscript. We appreciate your careful review.

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