

Response to Anonymous Referee #3 (RC2)

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Decadal transition in the source structure of PM_{2.5} at a coastal background site in North China: weakening combustion and nitrate amplification during high-PM_{2.5} conditions

Overall assessment. The manuscript addresses a relevant ACP question: how the composition and source-related characteristics of PM_{2.5} evolved at a regional-background receptor during China's clean-air-control period. The study contributes a long-term dataset from a location where continental transport, coastal processing, sea salt, and shipping-related influence can overlap. The combination of measured composition and PMF provides a convincing description of weakening combustion-related and sulfate signals and increasing nitrate importance.

My main recommendation is to keep the interpretation proportional to the observational design. The manuscript provides strong evidence for a stage-dependent compositional transition at Tuoji Island. It provides supporting thermodynamic context, but it does not independently quantify the effects of emission policy, meteorology, transport, or gas-phase precursor availability. The revised manuscript should distinguish these evidence levels clearly and moderate the corresponding causal claims. I recommend the paper for publication in ACP after the following specific comments are addressed:

Response. We appreciate the referee's positive assessment of the dataset and the source-apportionment results, and we found the recommendation to keep the interpretation proportional to the observational design particularly helpful. The revision follows this principle throughout. Causal language about emission controls has been moderated to associative wording (Comment 2), the thermodynamic outputs are now identified as related diagnostics derived from the same measured ion composition rather than as independent confirmation (Comment 3), sample-matched meteorology and a backward-trajectory climatology have been added to describe the transport setting of each stage (Comment 4), the distinction between the stage-average source structure and high-PM_{2.5} amplification is preserved in the title, Abstract, results, and Conclusions (Comment 5), and the Conclusions now close with a consolidated statement of the interpretation limits (Comment 6). The individual changes are detailed below.

Comment 1. Define the principal novelty more directly. The sulfate-to-nitrate transition has been reported in Beijing, Tianjin, and other North China studies cited by the authors. Sect. 3.3 already identifies the added value of the long-term coastal-background record; please make the same point explicitly in the Introduction so that the new contribution is clear before the results are presented.

Response. We agree that the contribution needed to be stated before the results. The revised Introduction now identifies two concrete additions to the urban evidence. First, it tests whether the sulfate-to-nitrate transition remains evident after continental

outflow has undergone transport, mixing, and coastal processing before reaching a regional background receptor. Second, it distinguishes the late-stage average source structure from the factors amplified during high-PM_{2.5} conditions. The trajectory climatology shows that continental pathways remained dominant across all stages and both high-PM_{2.5} subsets, placing the chemical transition in a predominantly continental transport setting. These points are now stated in the Introduction, developed in Sect. 3.3, and summarised in the Conclusions.

Revised manuscript text (Sect. 1; p. 2, lines 60-65): Urban records from North China have already documented declining combustion- and sulfate-related signals and an increasing relative importance of nitrate (Dai et al., 2023; Wang et al., 2022; Zhai et al., 2021). What remains less well constrained is whether the same transition persists at a regional background receptor over the full clean-air-action period, after continental outflow has undergone transport, mixing, and coastal processing.

Revised manuscript text (Sect. 1; p. 2, lines 65-75): Previous coastal studies have shown sea-salt displacement by nitrate, marine and combustion-related sources of reactive chlorine (Zong et al., 2026), and overlapping influences from shipping, continental outflow, and secondary formation (Li et al., 2018; Zhang et al., 2021; Wu et al., 2024). Tuoji Island, in the central Bohai Strait, lies at the intersection of North China continental pathways and marine inflow. The record tests whether the urban-observed transition is retained in processed regional aerosol and distinguishes the stage-average source structure from the factors amplified during high-PM_{2.5} conditions.

Comment 2. Moderate causal language concerning emission controls. The decline in EC, the combustion-related PMF factor, and sulfate is consistent with the timing and known direction of emission controls. However, the study does not perform a formal policy-attribution or meteorological-normalisation analysis. The current discussion is mostly careful, but the Abstract statement that emission reductions "reorganised" PM_{2.5} pollution is stronger than the direct evidence. Wording such as "during the emission-control period" or "consistent with the emission-reduction trajectory" would be more balanced.

Response. This point is well taken, and we have adopted the associative wording the referee suggested. The Abstract no longer states that emission reductions "reorganised" PM_{2.5} pollution; the opening sentence now says that clean-air actions "have coincided with" the observed changes, which claims temporal association rather than attribution. In Sect. 3.3, the comparison with the CAQIEI emission inventory (Kong et al., 2024) is framed with "consistent with", and the weakening of primary combustion is described as occurring during the period of sustained controls, matching the "during the emission-control period" formulation. No formal policy-attribution or meteorological-normalisation analysis is claimed anywhere in the revised text.

Revised manuscript text (Abstract; p. 1, lines 20-25): Long-term clean-air actions have coincided with substantial changes in fine-particle pollution in North China, but

long-term source transitions at regional background sites remain insufficiently constrained.

Revised manuscript text (Sect. 3.3; pp. 10-11, lines 345-355): The weakening of primary combustion occurred during sustained controls on coal combustion, boilers, key industries, residential coal use, and diesel vehicles (State Council of the People's Republic of China, 2013, 2018). ... These inventory trends are consistent with the weaker combustion signal observed at Tuoji Island.

Revised manuscript text (Sect. 3.3; p. 11, lines 355-360): This pattern is consistent with a faster decline in sulfate influence than in nitrate-related secondary aerosol.

Comment 3. Separate supporting thermodynamic diagnostics from independent lines of evidence. The increase in NO_3^- concentration and fraction and the nitrate-rich PMF factor are two related but useful forms of evidence. By contrast, NH_4^+ excess, reverse-mode ALWC, and model-resolved NH_4NO_3 all depend on the observed particle-phase ion composition; they strengthen the thermodynamic consistency of the interpretation but are not fully independent confirmation. Please state this relationship once in Sect. 3.3. In the same revision, replace "greater NH_4^+ availability" with a precise particle-phase stoichiometric description (positive NH_4^+ excess does not directly measure gaseous NH_3 availability), and describe ALWC as a model diagnostic associated with the late-stage aerosol state rather than as evidence that wetter particles caused the nitrate increase.

Response. All three requested changes have been made. First, Sect. 3.3 now states that the ion-balance indicators, reverse-mode ALWC, and stable-mode salt allocation are derived from the same particle-phase ion measurements. Their agreement describes related aspects of the same nitrate-rich aerosol state but does not provide independent confirmation of mechanism; Text S4 carries the same statement, and the model-resolved NH_4NO_3 allocation describes the salt distribution calculated from those same inputs. Second, "greater NH_4^+ availability" has been deleted. The interpretation now uses particle-phase stoichiometry, and Sects. 2.4 and 3.3 state directly that these quantities do not measure gas-phase NH_3 . Third, ALWC is described throughout as model-diagnosed, and its co-occurrence with elevated NO_3^- and NH_4^+ is presented as consistent with, not the cause of, NH_4NO_3 -related accumulation; the Abstract uses the same wording. Reverse-mode pH is used only to examine sensitivity to ion balance, not to infer ambient aerosol acidity or stage differences in acidity (Sect. 2.4 and Text S4; Song et al., 2018).

Revised manuscript text (Sect. 3.3; p. 11, lines 370-375): The transition from negative to positive particulate NH_4^+ excess indicates a shift in ionic stoichiometry after sulfate neutralisation that is consistent with a larger particulate ammonium-nitrate pool; it is not a measurement of gas-phase NH_3 availability.

Revised manuscript text (Sect. 3.3; p. 11, lines 370-385): The ISORROPIA-II metastable calculation provides a related thermodynamic diagnostic (Fig. 5c; Text S4, Table S6, and Fig. S7). Median model-diagnosed ALWC in late-stage high- $\text{PM}_{2.5}$

samples was $22.00 \mu\text{g m}^{-3}$, compared with 14.95 and $12.49 \mu\text{g m}^{-3}$ in the early and middle stages; median NO_3^- and NH_4^+ reached 21.40 and $7.02 \mu\text{g m}^{-3}$. Their co-occurrence is consistent with NH_4NO_3 -related accumulation under wetter particle conditions. However, the ion-balance indicators, reverse-mode ALWC, and stable-mode salt allocation are all derived from the same particle-phase ion measurements. Their agreement therefore describes a coherent nitrate-rich aerosol state but does not provide independent confirmation of the underlying mechanism. Because concurrent gaseous NH_3 , HNO_3 , and HCl were unavailable, these diagnostics are used for relative stage comparison rather than direct inference of gas–particle partitioning (Fountoukis and Nenes, 2007; Song et al., 2018).

Revised manuscript text (Sect. 2.4; p. 4, lines 140-145): The ratio describes particulate ammonium relative to that required for sulfate neutralisation, and NH_4^+ excess was calculated as $n(\text{NH}_4^+) - 2n(\text{SO}_4^{2-})$. These quantities are particle-phase stoichiometric indicators and do not represent gas-phase NH_3 .

Revised manuscript text (Abstract; p. 1, lines 30-35): Particle-phase ion stoichiometry and model-diagnosed aerosol liquid water were consistent with a late-stage shift toward NH_4NO_3 -related accumulation.

Revised Supplement text (Text S4; p. 11): ISORROPIA-II used particle-phase ions, relative humidity, and temperature in reverse mode under stable and metastable assumptions (Fountoukis and Nenes, 2007; Song et al., 2018). Metastable ALWC, stable-mode salts, and particle-phase ion-balance quantities all depend on the same measured ion set. They therefore describe related aspects of the same aerosol state and should be interpreted together rather than as independent diagnostics. ... Reverse-mode pH is sensitive to ion measurement errors and charge balance (Song et al., 2018). Here, it is used only to examine the model response to ion balance, not to infer ambient aerosol acidity or stage differences in acidity.

Comment 4. Acknowledge the possible influence of meteorology and transport. Add a concise statement in Sect. 3.3 explaining that changes in meteorology and regional transport may also contribute to the stage differences; where readily available, a brief supporting discussion of observed wind conditions would strengthen this context.

Response. Section 2.3 describes daily meteorology matched to the sampling dates and 72 h GDAS1-driven HYSPLIT trajectories (2652 trajectories for 663 dates). The early archive provides maximum-wind-speed directions for all 119 early-stage sampling dates; their most frequent directions were ESE (16.8 %) and WSW (12.6 %). The post-2017 archive provides daily direction records, so the two record types are tabulated separately (Table S9b). RH medians were 68.0 %, 62.1 %, and 74.5 %, and mean wind speeds were 4.34, 3.80, and 4.17 m s^{-1} across the stages (Table S9). Section 3.3 discusses these conditions together with the observed and equal-season-standardised trajectory frequencies.

Revised manuscript text (Sect. 2.3; p. 3, lines 115-120): Ground meteorological data were obtained from the Tuoji Island monitoring records, including the FRT FWS-

series automatic weather station. Daily air temperature, relative humidity, pressure, and wind speed were matched to PM_{2.5} sampling dates. Temperature and relative humidity were used in ISORROPIA-II diagnostics. Early-stage directions refer to the direction at maximum wind speed on 119 sampling dates. Middle- and late-stage directions are archived daily station directions with 242 and 271 valid matches, respectively. Frequencies for these two record types are reported separately in Table S9b.

Revised manuscript text (Sect. 2.3; pp. 3-4, lines 120-130): To characterise transport pathways, 72 h backward trajectories were calculated with the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015) for 663 unique sampling dates. ... The calculations used NOAA Global Data Assimilation System 1 (GDAS1) meteorological fields at 1° × 1° spatial and 3 h temporal resolution (NOAA Air Resources Laboratory, 2026). ... A four-cluster solution was adopted after solutions with two to eight clusters were compared using total spatial variance, silhouette score, cluster size, and geographical separation. Details are provided in Text S6, Table S10, and Fig. S9.

Revised manuscript text (Sect. 3.3; p. 10, lines 325-345): Sample-matched meteorology differed among stages (Table S9). In the full composition data set, median relative humidity was 68.0 %, 62.1 %, and 74.5 %, and mean wind speed was 4.34, 3.80, and 4.17 m s⁻¹ in the early, middle, and late stages, respectively. The early-stage maximum-wind directions were most frequently east-southeasterly (16.8 %) and west-southwesterly (12.6 %; Table S9b). Four trajectory clusters were resolved: far western/northwestern continental (20.4 %), regional mainland (36.4 %), northern/northwestern continental (21.8 %), and southeastern marine (21.4 %) pathways (Fig. S9). The marine pathway accounted for 50.3 % of summer trajectories and 3.2 % of winter trajectories. Its observed stage frequencies were 23.3 %, 22.3 %, and 19.7 %; after equal-season standardisation, they were 18.9 %, 26.0 %, and 20.4 %. The far western/northwestern pathway was less frequent in the early stage (14.5 %, 22.2 %, and 21.2 % across the three stages); the corresponding standardised frequencies were 16.1 %, 19.9 %, and 20.7 %. Regional-mainland frequencies also differed after standardisation (44.2 %, 33.5 %, and 37.2 %). Continental pathways together accounted for 76.7 %–80.3 % of trajectories across stages and predominated in both the component-based and PMF high-PM_{2.5} subsets. The higher late-stage humidity coincided with the rebound in stage-mean PM_{2.5}. The trajectory results place the sulfate-to-nitrate transition in a predominantly continental transport setting at this coastal background receptor.

Revised Supplement text (Text S6; p. 15): Daily meteorological records were matched to the full composition samples. Valid temperature, relative-humidity, and wind-speed records were available for 105, 242, and 271 samples across the three stages, corresponding to 88.2 %, 93.1 %, and 93.4 % coverage (Table S9). Wind-direction frequencies were summarised in 16 compass sectors. Early-stage directions at maximum wind speed and middle- and late-stage daily directions were tabulated

separately. The meteorological summaries in the manuscript use the full composition sample set.

Comment 5. Distinguish late-stage average conditions from high-PM_{2.5} amplification. In 2021–2023, the port-related factor is the largest stage-average PMF factor, whereas secondary nitrate becomes the largest amplified factor in the high-PM_{2.5} subset. The title, abstract, and conclusion should preserve this distinction. "Increasing nitrate importance" or "nitrate amplification during high-PM_{2.5} conditions" is more precise than a general statement that all late-stage pollution is nitrate-driven.

Response. We fully agree and have adopted the referee's suggested phrase. The title now reads "nitrate amplification during high-PM_{2.5} conditions", and the distinction between the stage-average source structure and the amplified high-PM_{2.5} subset is preserved in four places: the title, the Abstract, Sect. 3.2, and the Conclusions. In each of these, the port-related mixed factor is identified as the largest 2021–2023 stage-average factor (27.1 %), while secondary nitrate is described as the factor amplified in the stage-relative high-PM_{2.5} subset (27.7 %). Nowhere does the revised text state that late-stage pollution as a whole is nitrate-driven.

Revised manuscript text (Title; p. 1): Decadal transition in the source structure of PM_{2.5} at a coastal background site in North China: weakening combustion and nitrate amplification during high-PM_{2.5} conditions

Revised manuscript text (Abstract; p. 1, lines 25-35): In 2021–2023, the port-related mixed factor was the largest stage-average PMF factor (27.1 %), whereas secondary nitrate was selectively amplified in stage-relative high-PM_{2.5} samples, reaching 27.7 % and coinciding with a NO₃[−] fraction of 19.2 %. ... The record therefore shows weaker combustion and sulfate signals but stronger nitrate amplification during late-stage high-PM_{2.5} conditions, while distinguishing this polluted-subset behaviour from the stage-average source structure.

Revised manuscript text (Sect. 3.2; p. 9, lines 300-315): The stage-relative high-PM_{2.5} PMF comparison used 11, 17, and 35 samples remaining after mineral-dust-outlier removal from the PMF subset in 2012, 2019–2020, and 2021–2023, respectively (Fig. 4; Table S8a). In 2012, the mixed combustion, secondary sulfate, and secondary nitrate factors contributed 47.4 %, 27.5 %, and 7.6 %. In 2019–2020, secondary nitrate increased to 21.5 %, comparable with secondary sulfate (20.4 %), the mixed combustion factor (19.9 %), and the port-related mixed factor (19.9 %). By 2021–2023, secondary nitrate reached 27.7 %, exceeding the port-related mixed (19.2 %), secondary sulfate (18.0 %), and mixed combustion (17.0 %) factors; relative to the screened stage-average contribution of 16.4 %, its enrichment ratio was 1.69. Because the early-stage high-PM_{2.5} subset contained 11 samples, the comparison emphasises factor ranking and broad stage differences. Thus, the late-stage high-value subset was nitrate-amplified, whereas the port-related mixed factor had the largest contribution to the late-stage average.

Revised manuscript text (Sect. 4; p. 13, lines 405-415): The late-stage PMF results differed between the stage average and the stage-relative high-PM_{2.5} subset. In all 2021–2023 PMF samples, the port-related mixed factor was the largest factor at 27.1 %, while the mixed biomass-burning and coal-combustion factor had declined from 43.4 % in 2012 to 13.7 %. In the stage-relative high-PM_{2.5} subset, secondary nitrate was instead the dominant amplified factor, increasing to 27.7 % and coinciding with a NO₃[−] fraction of 19.2 %. The late-stage average and high-value subset therefore represent distinct source-composition patterns.

Comment 6. Improve the balance between results and limitations. The conclusion repeats many values already presented in Sect. 3.1–3.3. Condense these repeated results and add a concise paragraph stating the main interpretation limits: non-continuous stage coverage (including COVID-affected sampling in 2020 and differing seasonal coverage among stages), the smaller PMF subset, and the diagnostic nature of the reverse-mode thermodynamic results.

Response. The Conclusions have been restructured along the requested lines. The repeated numerical detail has been condensed so that each headline value appears once, and a dedicated limitations paragraph now covers every point the referee listed: the non-contiguous, unequal PMF windows (defined in Sect. 2.5) and their differing seasonal coverage; the COVID-19-affected year 2020 within the middle period, together with the sensitivity result obtained when that year is excluded (Text S5 and Table S8c report that the middle-period balance between the port-related mixed and secondary nitrate factors changes); the small size of the PMF high-PM_{2.5} subsets, with only 11 early-stage samples; the assumption that each factor has a broadly stable chemical composition across periods; and the diagnostic, non-independent nature of the reverse-mode thermodynamic results, which cannot distinguish precursor pathways because gaseous precursors and organic molecular tracers were unavailable. The diagnostic status of the thermodynamic quantities is also restated where the Conclusions summarise the ion-balance results.

Revised manuscript text (Sect. 4; p. 13, lines 420-430): Observations were unavailable during 2014–2016. The PMF comparisons are used primarily to evaluate the direction of stage changes. The three PMF windows were unequal and differed in seasonal coverage, and the early high-PM_{2.5} subset contained 11 samples. The middle period included the COVID-19-affected year 2020 (Liu et al., 2022); excluding that year changed the balance between the port-related mixed and secondary nitrate factors (Text S5 and Table S8c). The pooled PMF assumes that the chemical composition of each factor remains broadly stable across periods (Xie et al., 2022). The thermodynamic calculations assess consistency with ammonium-nitrate formation; they do not distinguish among precursor pathways because gaseous precursors and organic molecular tracers were unavailable.

Revised manuscript text (Sect. 4; p. 13, lines 410-420): Taken together, these related diagnostics support the high-value nitrate signal without independently resolving its formation mechanism.

Revised manuscript text (Sect. 2.5; p. 5, lines 170-180): Only samples with all common PMF input species and corresponding uncertainties were retained, giving 54 samples for 2012, 81 for 2019–2020, and 178 for 2021–2023 (313 in total); the corresponding stage-relative high-PM_{2.5} subsets contained 11, 17, and 35 samples (Text S5 and Table S8a). ... These non-contiguous windows of unequal duration were used for stage-level comparisons.

Revised Supplement text (Text S5; p. 14): A sensitivity comparison excluding 2020 retained the decrease in the mixed combustion factor and the increase in secondary nitrate, but it changed the quantitative middle-period source balance. For all middle-period PMF samples, the port-related mixed factor decreased from 26.4 % in 2019–2020 to 15.6 % in 2019 alone, while secondary nitrate increased from 17.2 % to 23.6 %.

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