

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. This manuscript presents a 12-year (2012–2023) record of PM_{2.5} chemical composition and source apportionment at Tuoji Island, a coastal background site in the Bohai Strait. The integration of filter-based chemical characterization, positive matrix factorization (PMF), ion-balance diagnostics, and ISORROPIA-II thermodynamic modeling is well aligned with the scope of ACP. The long observation period at a regional-background setting, where continental transport, coastal processing, sea salt, and shipping influence can overlap, distinguishes the study from urban or episode-based source-apportionment work. However, the scientific contribution of the manuscript is constrained by a fundamental problem of novelty differentiation. The core conclusion that a transition from combustion- and sulfate-dominated pollution toward nitrate-driven secondary pollution under China's clean-air actions, has been extensively reported in urban North China (Zhai et al., 2021, Nat. Geosci.; Dai et al., 2023; Wang et al., 2022; Chu et al., 2023). The manuscript does not sufficiently articulate or demonstrate what the background-site perspective adds beyond confirming urban trends at a different location. In addition, several issues needed to be clarified. I recommend the paper for publication in ACP after substantial revision addressing the following specific comments.

Response. We thank the referee for this careful and constructive assessment, and in particular for identifying novelty differentiation as the central issue. The revised manuscript now defines the background-site contribution in concrete terms: it tests whether the urban-observed sulfate-to-nitrate transition remains evident in processed continental outflow at a regional receptor, and it separates the late-stage average source structure from the factors amplified during high-PM_{2.5} conditions. A backward-trajectory climatology (2652 HYSPLIT trajectories for 663 sampling dates) shows that continental pathways dominated every stage and both high-PM_{2.5} subsets. Seasonally standardised comparisons within the regional-mainland pathway further show declining EC and an increasing NO₃⁻/SO₄²⁻ ratio (Table S11a). The title has been narrowed to the single-site scope, and the stage definitions, PMF sample coverage, thermodynamic diagnostics, mixed PMF factors, organic-speciation gap, and policy statements have been revised as requested. Point-by-point responses to the nine specific comments and three technical corrections follow.

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. ... the

chemical components or source factors preferentially amplified during later-stage high-PM_{2.5} conditions remain uncertain.

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.

Specific comment 1. The sulfate-to-nitrate transition has been documented in Beijing (Wang et al., 2022), Tianjin (Dai et al., 2023), and the BTH "2+26" cities (Chu et al., 2023), and mechanistically explained by Zhai et al. (2021). Sect. 3.3 identifies the added value of the long-term coastal-background record, but the Introduction does not state explicitly what remains unknown after these studies and how the Tuoji Island record fills those gaps. Please articulate the specific quantitative insight that the background site provides, for example, the relative magnitude of regional transport versus local formation, or the background-level compositional response to emission controls, and state this contribution in the Introduction before the results are presented.

Response. We compared chemistry within individual trajectory clusters, with equal weighting of the four seasons. Within the regional-mainland pathway C2, NO₃⁻ concentrations were 7.21, 6.65, and 12.58 μg m⁻³, EC concentrations were 1.67, 1.56, and 0.96 μg m⁻³, and NO₃⁻/SO₄²⁻ ratios were 0.61, 1.50, and 2.34 across the three stages (Table S11a). The compositional change therefore remains evident within a common continental pathway reaching the offshore receptor after seasonal sampling differences are accounted for. This provides evidence of changing regional aerosol composition beyond a comparison of the proportions of continental and marine air masses. The Introduction now states this within-pathway comparison explicitly. Section 3.3 and the Conclusions report its results and distinguish the late-stage average source structure from nitrate amplification in high-PM_{2.5} samples.

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. ... the chemical components or source factors preferentially amplified during later-stage high-PM_{2.5} conditions remain uncertain.

Revised manuscript text (Sect. 1; p. 2, lines 75-80): We ask three questions: (1) did the source structure of PM_{2.5} change across the observed stages even though stage-mean mass did not decline monotonically? (2) within stage-relative high-PM_{2.5}

samples selected after mineral-dust-outlier removal, which chemical component or PMF-resolved factor was preferentially amplified? (3) were the observed nitrate changes accompanied by consistent shifts in particle-phase ion stoichiometry and model-diagnosed aerosol liquid water?

Revised manuscript text (Sect. 3.3; pp. 10-11, lines 340-370): Composition also changed within the regional-mainland pathway (C2). After mineral-dust screening and equal-season standardisation, NO_3^- concentrations were 7.21, 6.65, and $12.58 \mu\text{g m}^{-3}$ across the three stages, EC concentrations were 1.67, 1.56, and $0.96 \mu\text{g m}^{-3}$, and $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios were 0.61, 1.50, and 2.34 (Table S11a). Thus, weaker combustion signals and greater nitrate importance were observed within the same broad continental pathway after seasonal sampling differences were accounted for. ... Among late-stage high- $\text{PM}_{2.5}$ samples, the pathway-weighted NO_3^- fraction was 22.9 % for C2 and 12.9 % for the marine pathway; the C2-weighted secondary-nitrate PMF contribution was 29.6 %, compared with 19.9 % for all screened late-stage PMF samples weighted for C2 (Tables S11b–S11c). Autumn and winter high- $\text{PM}_{2.5}$ samples had NO_3^- fractions of 22.8 % and 26.1 % ($n = 14$ and 15), compared with 13.5 % in spring ($n = 26$; Table S11d).

Specific comment 2. The 2014–2016 gap means the record is not continuous, and the three stages (2012–2013, 2017–2020, 2021–2023) appear to be defined by data availability rather than by scientific or policy milestones. The 2012–2013 stage contains only ~18 months of data. Please discuss how this discontinuity affects trend interpretation, whether the stage boundaries correspond to identifiable inflection points in emission inventories, and acknowledge the non-continuous coverage, including COVID-affected sampling in 2020 and differing seasonal coverage among stages, as a limitation in the Conclusion.

Response. This point is well taken, and it also led us to correct an internal inconsistency. The original Sect. 2.3 attributed the 2014–2016 gap to insufficient samples, which contradicted the sampling windows stated in Sect. 2.1. Section 2.1 now states plainly that observations were interrupted between the two sampling windows, so no samples exist for 2014–2016, and Sect. 2.3 states that this gap precluded a separate stage comparison. On the stage boundaries, we now describe their basis honestly: they follow observation continuity, temporal coverage, and the policy context of the two clean-air-action phases. They correspond broadly to the two action periods represented in the CAQIEI emission inventory (Kong et al., 2024), but they were not fitted to inventory inflection points, and we do not claim that they mark emission turning points. Consistent with this, Sect. 2.5 states that the non-contiguous windows of unequal duration are used for stage-level comparisons rather than continuous annual trend analysis, so no trend is interpolated across the gap. The Conclusions now carry the requested limitations explicitly: the three PMF windows were unequal and differed in seasonal coverage, the early high- $\text{PM}_{2.5}$ subset contained only 11 samples, and the middle period included the COVID-19-affected

year 2020, with the effect of excluding that year reported as a sensitivity result (Text S5 and Table S8c).

Revised manuscript text (Sect. 2.1; p. 3, lines 90-100): Samples were collected with a high-volume sampler (Hi-Vol 3000, Ecotech Company, Australia) on quartz-fibre filters (Whatman QM-A, 8×10 in.) at $1.13 \text{ m}^3 \text{ min}^{-1}$ for 24 h, nominally every 3 d. Filters were pre-baked at 500°C for 4 h. The observation record comprises March 2012–August 2013 and September 2017–February 2024; observations were interrupted between these windows, so no samples were available during 2014–2016. After sample-validity and component-completeness checks, 669 samples were retained for stage-level statistics.

Revised manuscript text (Sect. 2.3; p. 4, lines 130-140): The three analysis periods were defined from observation continuity, temporal coverage, and the policy context of regional air-pollution control. The 2012–2013 period represents the pre-action background state. The 2014–2016 observation gap precluded a separate stage comparison. The 2017–2020 period covers the further strengthening of clean-air controls, and 2021–2023 follows completion of the Three-Year Action Plan. The stages follow the available observation windows and correspond broadly to the two clean-air-action phases represented in the CAQIEI inventory (Kong et al., 2024). They contain 119, 260, and 290 valid samples. After mineral-dust-outlier screening, the highest 20 % of $\text{PM}_{2.5}$ values were selected independently within each stage and are termed stage-relative high- $\text{PM}_{2.5}$ samples (Sect. 2.6).

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- $\text{PM}_{2.5}$ 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.

Specific comment 3. The stage-level composition analysis uses 672 valid samples, whereas PMF uses 313 samples with complete common inputs, and the PMF comparison periods (2012; 2019–2020; 2021–2023) are narrower and of unequal length relative to the composition stages. Sect. 2.5 explains why the two datasets differ, but the main component changes in the PMF subset should be confirmed as consistent with those in the full dataset. Please state the three PMF-period sample counts explicitly, comment on the comparability of the unequal-length comparison periods (a single early year versus multi-year later periods), and note this mismatch as a limitation where quantitative factor comparisons are discussed.

Response. We appreciate this request, which was also raised by Referee #1, and all three parts have been implemented. First, Sect. 2.5 now states the PMF-period sample counts explicitly: 54 samples for 2012, 81 for 2019–2020, and 178 for 2021–2023 (313 in total), with high-PM_{2.5} subsets of 11, 17, and 35 samples. Second, Sect. 2.5 now characterises these windows as non-contiguous and of unequal duration and uses them for stage-level comparisons; Sect. 3.2 correspondingly limits the high-PM_{2.5} comparison to factor ranking and broad stage differences because the early subset contains only 11 samples. Third, a new consistency check (Text S5, Tables S8a–S8b) confirms that the main component changes in the PMF subset agree with all composition samples from the same PMF-covered years (2012, 2019–2020, and 2021–2023; $n = 77, 110, \text{ and } 290$): six of seven indicators (PM_{2.5}, OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, NO₃⁻/SO₄²⁻) change in the same direction in each adjacent-stage contrast, with NH₄⁺ the only exception, showing small non-monotonic changes consistent with Sect. 3.1, and all seven agree from the early to the late stage. The mismatch is also recorded as a limitation in the Conclusions, quoted in our response to Specific comment 2. The revised composition analysis contains 669 valid samples after exclusion of three records without usable chemical concentrations; the PMF matrix remains unchanged at 313 samples.

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). A pooled solution provided a common factor-profile framework across periods, consistent with a previous PMF application that combined multiple sites and time periods (Yang et al., 2023). These non-contiguous windows of unequal duration were used for stage-level comparisons.

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). ... Because the early-stage high-PM_{2.5} subset contained 11 samples, the comparison emphasises factor ranking and broad stage differences.

Revised manuscript text (Sect. 3.2; p. 9, lines 310-320): The direction of change between adjacent stages agreed between the composition samples from the PMF-covered years (2012, 2019–2020, and 2021–2023) and the PMF subset for six of seven indicators; NH₄⁺ was the exception, with small, non-monotonic changes consistent with Sect. 3.1 (Text S5 and Table S8b). The main factor patterns were also retained in the $F_{\text{peak}} = 0.5$ sensitivity solution (Table S3b and Fig. S3). Excluding 2020 retained the decline in mixed combustion and the increase in secondary nitrate, but changed the quantitative middle-period balance between the port-related mixed and secondary nitrate factors (Text S5 and Table S8c).

Revised Supplement text (Text S5; p. 14): The 313-sample PMF matrix comprised 54 samples from 2012, 81 from 2019–2020, and 178 from 2021–2023; the corresponding

stage-relative high-PM_{2.5} PMF subsets contained 11, 17, and 35 samples (Table S8a). The PMF windows are unequal and non-contiguous and are used for stage contrasts. To assess subset representativeness, stage changes in PM_{2.5}, OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, and the NO₃⁻/SO₄²⁻ ratio were compared between all composition samples from the PMF-covered years (2012, 2019–2020, and 2021–2023; n = 77, 110, and 290) and the PMF subset. Six of seven indicators had the same direction in each adjacent-stage contrast; NH₄⁺ was the exception, with small changes of opposite sign consistent with its non-monotonic behaviour in the main text. All seven indicators agreed from the early to late stage (Table S8b).

Specific comment 4. The manuscript already states that particle-phase stoichiometry does not represent gas-phase NH₃ and that pH is a model diagnostic, this is appropriate. However, NH₄⁺ excess, reverse-mode ALWC, and model-resolved NH₄NO₃ all depend on the observed particle-phase ion composition; they support the internal consistency of a nitrate-rich aerosol state but are not fully independent confirmation of formation rates, gas–particle partitioning, or persistence. Without synchronous gas-phase NH₃, HNO₃, or HCl measurements, the gas–particle partitioning claims for NH₄NO₃ cannot be directly verified. Please state this relationship explicitly in Sect. 3.3, describe ALWC as a model diagnostic associated with the late-stage aerosol state rather than as evidence that wetter particles caused the nitrate increase, and replace "greater NH₄⁺ availability" with a precise particle-phase stoichiometric description.

Response. We fully agree with this characterisation, and all three requested changes have been made. Section 3.3 now states that the ion-balance indicators, reverse-mode ALWC, and stable-mode salt allocation all derive from the same particle-phase ion measurements. Their agreement describes a coherent nitrate-rich aerosol state but does not provide independent confirmation of formation rates, gas–particle partitioning, or persistence. Without concurrent gaseous NH₃, HNO₃, and HCl, the diagnostics are used for relative stage comparison. ALWC is described throughout as model-diagnosed, and its co-occurrence with elevated NO₃⁻ and NH₄⁺ is expressed as consistent with, not the cause of, NH₄NO₃-related accumulation. The phrase "greater NH₄⁺ availability" has been removed and replaced by a particle-phase stoichiometric description. Text S4 uses the same framing. 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. 2.4; p. 4, lines 140-145): The ratio describes particulate ammonium relative to that required for sulfate neutralisation, and NH₄⁺ excess was calculated as n(NH₄⁺) – 2n(SO₄²⁻). These quantities are particle-phase stoichiometric indicators and do not represent gas-phase NH₃.

Revised manuscript text (Sect. 3.3; p. 11, lines 370-375): The transition from negative to positive particulate NH₄⁺ 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 NH₃ availability.

Revised manuscript text (Sect. 3.3; p. 11, lines 375-385): 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. 3.3; p. 12, lines 385-390): This allocation shares the ion inputs used for the other thermodynamic diagnostics and describes another aspect of the same inorganic-ion shift.

Revised Supplement text (Text S4; p. 11): 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. Concurrent gaseous NH_3 , HNO_3 , and HCl were unavailable, so these outputs are used for relative stage comparison rather than as direct gas-particle partitioning evidence. 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.

Specific comment 5. Stage-mean $\text{PM}_{2.5}$ did not decrease monotonically ($51.75 \rightarrow 47.31 \rightarrow 54.41 \mu\text{g}/\text{m}^3$), with the 2021–2023 mean actually exceeding the 2012–2013 baseline. The manuscript attributes this to compositional changes, but meteorological variability between stages is not systematically addressed. Changes in synoptic circulation, boundary-layer height, and regional transport patterns may also contribute to the stage differences. Please add a concise discussion of meteorological influence in Sect. 3.3, to strengthen the attribution of late-stage $\text{PM}_{2.5}$ levels to emission-driven compositional shifts.

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.

Boundary-layer height observations were unavailable at Tuoji Island during the sampling periods. The meteorological discussion uses the available surface observations and backward trajectories to describe the stage-specific conditions.

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 manuscript text (Sect. 4; p. 13, lines 415-425): The late stage was more humid in the full sample data set. The four-cluster trajectory climatology identified both continental and marine air masses, with continental pathways dominant in all stages and in stage-relative high-PM_{2.5} samples. Seasonally standardised frequencies differed among stages, particularly for the regional-mainland and marine pathways. Within the regional-mainland pathway, equal-season-standardised NO₃⁻ concentrations increased from 7.21 to 12.58 µg m⁻³ between the early and late stages, while EC decreased from 1.67 to 0.96 µg m⁻³. This extends the urban evidence to compositional changes within a shared continental pathway reaching the coastal background receptor.

Specific comment 6. The manuscript repeatedly invokes "regional transport" as a key process at this coastal background site, yet no backward trajectory analysis (HYSPLIT, FLEXPART), PSCF/CWT analysis, or concentration-weighted trajectory method is presented. Without this, the transport narrative remains unsubstantiated. At minimum, a seasonal trajectory climatology distinguishing continental outflow from marine/clean air masses would help contextualize the stage-level compositional changes and strengthen the background-site framing that is central to the manuscript's novelty claim.

Response. We added a seasonal backward-trajectory analysis. Section 2.3 describes 72 h HYSPLIT trajectories (Stein et al., 2015) for the 663 unique dates represented by 669 valid filter samples. Trajectories arrive at 500 m above ground level at 00:00, 06:00, 12:00, and 18:00 UTC, giving 2652 trajectories. The calculation uses NOAA

GDAS1 fields at $1^\circ \times 1^\circ$ and 3 h resolution, followed by Ward minimum-variance agglomerative clustering. Two- to eight-cluster solutions were compared using TSV, silhouette score, minimum cluster size, and geographical separation (Table S10). Four clusters first separate the southeastern marine pathway while retaining at least 541 trajectories in each cluster. The marine pathway accounts for 50.3 % of summer trajectories and 3.2 % of winter trajectories. The three continental pathways account for 76.7 %–80.3 % of all trajectories across stages, 84.2 %–96.6 % in the component-based high-PM_{2.5} subset, and 83.6 %–91.2 % in the PMF high-PM_{2.5} subset. Observed and equal-season-standardised frequencies are reported in Sect. 3.3 and Text S6, with the mean paths and frequencies shown in Fig. S9. The higher late-stage humidity coincided with the rebound in stage-mean PM_{2.5}. Seasonally standardised pathway frequencies differed among stages, especially for regional-mainland and marine pathways. Continental pathways nevertheless predominated in every stage and in the component-based and PMF high-PM_{2.5} subsets, placing the chemical transition in a predominantly continental transport setting at this coastal background receptor. We also compared chemistry and PMF results conditional on each trajectory class; methods are in Text S6 and results in Tables S11a–S11d.

Revised manuscript text (Sect. 2.3; pp. 3-4, lines 120-135): 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. These dates represent 669 filters because six dates contained duplicate filters. Trajectories were initiated at 500 m above ground level, an arrival height used in previous regional-background and North China aerosol studies (Zong et al., 2016; Zhang et al., 2018). Four arrival times per sampling date (00:00, 06:00, 12:00, and 18:00 UTC) represented airflow during the 24 h filter-sampling period, giving 2652 trajectories. The calculations used NOAA Global Data Assimilation System 1 (GDAS1) meteorological fields at $1^\circ \times 1^\circ$ spatial and 3 h temporal resolution (NOAA Air Resources Laboratory, 2026). The trajectories were grouped using Ward minimum-variance agglomerative clustering. For comparison, stage frequencies were also calculated with equal weighting of the four seasons. 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. To compare composition under similar transport pathways, chemical concentrations and PMF contributions were weighted by each cluster's share of the four trajectories on the sampling date. Stage comparisons of chemical composition gave equal weight to the four seasons (Text S6 and Tables S11a–S11d).

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): Solutions with two to eight clusters were compared using the change in total spatial variance (TSV), silhouette score, minimum cluster size, and geographical separation (Table S10). Previous HYSPLIT studies have used TSV together with the geographical separation of mean pathways to select the cluster number (Hong et al., 2022; Jaén et al., 2025). The two-cluster solution had the highest silhouette score (0.430), but merging from three to two clusters increased TSV by 30.0 % and combined distinct pathways. Four clusters were selected because this was the first solution that separated the southeastern marine pathway and all four clusters contained at least 541 trajectories. The five-cluster solution further divided the far western/northwestern pathway, producing a small subgroup of 129 trajectories, and had a lower silhouette score (0.228). Stage and seasonal frequencies were calculated from all 2652 individual trajectories. Equal-season-standardised stage frequencies were calculated as the arithmetic mean of the four season-specific frequencies within each stage.

Revised Supplement text (Text S6; pp. 15-16): The regional-mainland cluster was most frequent (36.4 %), followed by the northern/northwestern continental (21.8 %), southeastern marine (21.4 %), and far western/northwestern continental (20.4 %) clusters (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 % in 2012–2013, 2017–2020, and 2021–2023, respectively; after equal-season standardisation, the corresponding frequencies were 18.9 %, 26.0 %, and 20.4 %. The lower early-stage frequency after standardisation reflects the larger summer share in the 2012–2013 sampling window. The far western/northwestern pathway occurred at frequencies of 14.5 %, 22.2 %, and 21.2 %, respectively, and 16.1 %, 19.9 %, and 20.7 % after standardisation. Standardised regional-mainland frequencies were 44.2 %, 33.5 %, and 37.2 %. Thus, differences in pathway frequency persisted after accounting for seasonal sampling. The three continental pathways together accounted for 76.7 %–80.3 % of all trajectories across stages, 84.2 %–96.6 % of trajectories associated with component-based high-PM_{2.5} samples, and 83.6 %–

91.2 % of those associated with the PMF high-PM_{2.5} subset. Continental transport predominated in each comparison.

Revised Supplement text (Fig. S9; p. 18): Figure S9. Four-cluster 72 h backward-trajectory climatology for Tuoji Island sampling dates using GDAS1 meteorological fields. (a) Mean paths and overall frequencies for all 2652 trajectories; the star marks Tuoji Island. (b) Observed stage frequencies. (c) Seasonal frequencies. Frequencies are based on individual trajectories rather than daily modal assignments; equal-season-standardised stage frequencies are reported in Text S6. No uncertainty intervals are shown.

Specific comment 7. The port-emission factor accounts for 26–27% of PM_{2.5} in the later stages, an unusually high contribution for a background site. The factor is identified by Na⁺, Fe, Ni, and Cu, but these species can also originate from sea salt, crustal material, or other industrial sources. While the manuscript acknowledges that this factor combines port activity, shipping, coastal aerosol, and regional metal inputs, the mixed character should be stated consistently in the Methods, Results, and figure captions. Please provide more rigorous factor validation, including V/Ni ratios, enrichment-factor analysis, or comparison with shipping emission inventories for the Bohai Sea region, and interpret the quantitative contribution cautiously given the factor's mixed nature.

Response. We are grateful for the concrete validation options offered here and have adopted the first of them. The V/Ni ratio of the factor profile is now reported in Sect. 3.2: it is 0.10, which is inconsistent with a pure ship-exhaust signature, and this diagnostic is the basis for interpreting the factor as mixed. Accordingly, the factor has been renamed the port-related mixed factor and this name and its mixed interpretation (port and shipping activity, ship-fuel combustion, coastal aerosol, and regional metal inputs) are now used consistently across the Abstract, Sect. 3.2, the Conclusions, and the supplementary methods and figure material (Text S2, Fig. S3, and Table S8c). A dedicated paragraph in Sect. 3.2 further cautions that this factor characterises the receptor setting and regional inputs and does not by itself explain the nitrate trends, and Sect. 2.5 restricts the interpretation of all metal-related factors to tracer profiles and broad stage patterns rather than small contribution differences, which is the cautious quantitative reading the referee requests. Of the two remaining options, an enrichment-factor analysis cannot be computed reliably for this data set because the crustal reference elements it requires (Al, Si, Ti) were not measured, a gap now stated openly in Sect. 2.2. A comparison with Bohai Sea shipping emission inventories was not undertaken: the referee's list presents the three approaches as alternatives, and the V/Ni diagnostic together with the profile composition already answers the validation question in the direction of caution, namely that the factor must not be read as pure ship exhaust.

Revised manuscript text (Sect. 3.2; pp. 8-9, lines 285-290): F6 contained Na⁺, Fe, Ni, and Cu but had a profile V/Ni ratio of 0.10, inconsistent with a pure ship-exhaust

signature; it was therefore interpreted as a mixed factor reflecting port and shipping activity, coastal aerosol, and regional metal inputs.

Revised manuscript text (Sect. 3.2; p. 9, lines 295-305): The port-related mixed factor combines port and shipping activity, ship-fuel combustion, coastal aerosol, and regional metal inputs, while the industrial-metal factor records trace-metal signals. The low V/Ni ratio and broad metal profile do not support interpreting F6 as pure ship exhaust. These factors help characterise the receptor setting and regional inputs but do not alone explain the increasing $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio or the late-stage nitrate enrichment.

Revised manuscript text (Sect. 2.5; p. 5, lines 180-185): The metal-related factors are therefore interpreted mainly from their tracer profiles and broad stage patterns rather than from small differences in contribution percentages.

Revised manuscript text (Sect. 2.2; p. 3, lines 110-115): Al, Si, Ti, and some refractory components were not measured, and OC represents carbon rather than total organic matter.

Revised Supplement text (Text S2; p. 7): F6 is characterised by Na^+ , Fe, Ni, and Cu, with 84.2 % of Na^+ , 50.7 % of Cu, 38.0 % of Ni, and 36.3 % of Fe assigned to the factor. Its V/Ni ratio is 0.10, which does not support a pure ship-exhaust interpretation. Considering the nearby shipping channels and ports, F6 is named the port-related mixed factor and represents overlapping port and shipping activity, ship-fuel combustion, coastal aerosol, and regional metal inputs (Liu et al., 2022; Jang et al., 2023).

Revised Supplement text (Fig. S3; p. 8): F2 and F6 are interpreted as the mixed biomass-burning and coal-combustion factor and the port-related mixed factor, respectively.

Specific comment 8. Only OC and EC are measured; no organic molecular markers, SOA tracers, or online aerosol mass spectrometry data are available. Given that the "secondary nitrate" PMF factor may co-vary with secondary organic aerosol, and that organic nitrate formation could contribute to the observed nitrate enhancement, the absence of organic speciation represents a significant interpretive gap. Please add a concise statement in Sect. 3.3 or the Conclusion discussing this limitation and its potential implications for the source-apportionment results.

Response. We agree that the absence of organic speciation limits the interpretation, and the requested statements have been added in three places. Section 2.2 now records the measurement scope explicitly, including that OC represents carbon rather than total organic matter. The closing paragraph of Sect. 3.3 states that the available ion stoichiometry supports an interpretation dominated by inorganic ammonium nitrate, while the absence of organic molecular tracers prevents assessment of co-varying secondary organic aerosol and organic-nitrate processes, which is precisely the interpretive gap the referee describes: we can constrain the inorganic pathway but cannot exclude a co-varying organic contribution. The Conclusions carry the same limitation for the source-apportionment and thermodynamic results, stating that the

calculations do not distinguish among precursor pathways because gaseous precursors and organic molecular tracers were unavailable.

Revised manuscript text (Sect. 2.2; p. 3, lines 110-115): Al, Si, Ti, and some refractory components were not measured, and OC represents carbon rather than total organic matter.

Revised manuscript text (Sect. 3.3; p. 13, lines 395-400): The available ion stoichiometry supports an interpretation dominated by inorganic ammonium nitrate, while the absence of organic molecular tracers prevents assessment of co-varying secondary organic aerosol and organic-nitrate processes.

Revised manuscript text (Sect. 4; p. 13, lines 425-430): 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.

Specific comment 9. The conclusion that "coordinated control of NO_x, NH₃, and VOCs" is needed has been stated in numerous prior studies. The study does not perform a formal policy-attribution analysis, so the policy recommendation should be proportional to the evidence. Please leverage the specific background-site perspective to provide more targeted recommendations, for example, quantifying the relative importance of regional transport versus local formation, or identifying which precursor controls would yield the greatest benefit under the observed late-stage thermodynamic conditions, rather than restating generic emission-control conclusions.

Response. We added pathway-conditioned chemistry and PMF comparisons and seasonal statistics to make the recommendation specific to the coastal-background observations. We compared chemistry within individual trajectory clusters, with equal weighting of the four seasons. Within the regional-mainland pathway C2, NO₃⁻ concentrations were 7.21, 6.65, and 12.58 μg m⁻³, EC concentrations were 1.67, 1.56, and 0.96 μg m⁻³, and NO₃⁻/SO₄²⁻ ratios were 0.61, 1.50, and 2.34 across the three stages (Table S11a). The compositional change therefore remains evident within a common continental pathway reaching the offshore receptor after seasonal sampling differences are accounted for. This provides evidence of changing regional aerosol composition beyond a comparison of the proportions of continental and marine air masses. In late-stage high-PM_{2.5} samples, the C2-weighted NO₃⁻ fraction was 22.9 %, compared with 12.9 % for the marine pathway. The C2-weighted secondary-nitrate PMF contribution was 29.6 %, compared with 19.9 % in all screened late-stage PMF samples weighted for C2 (Tables S11b–S11c). Autumn and winter high-value samples had NO₃⁻ fractions of 22.8 % and 26.1 % (n = 14 and 15), compared with 13.5 % in spring (n = 26; Table S11d). These results support regional secondary-nitrate control under continental-influenced high-PM_{2.5} conditions and a particular focus on autumn and winter nitrate-rich samples. We removed the generalisation that all high-value conditions were humid: the late-stage high-value RH median was 68.3 %, compared with 75.0 % for all screened late-stage samples with valid

meteorology. The revised recommendation in the Conclusions follows these pathway and seasonal observations.

Revised manuscript text (Sect. 4; p. 13, lines 430-435): The coastal-background evidence supports prioritising regional secondary-nitrate control in continental-influenced high-PM_{2.5} conditions, alongside continued reductions in primary emissions. The higher nitrate fractions in autumn and winter high-PM_{2.5} samples identify these seasons as priorities for coordinated management of NO_x and NH₃ in upwind areas.

Technical correction 1. Revise the title to reflect the single-site design. "Over North China" may be read as a multi-site regional analysis. Replace it with wording such as "at a coastal background site in North China" to match the spatial scope of the evidence.

Response. Agreed, and the change has been made using the wording the referee suggests. Referee #1 raised the same point, and the two requests were resolved with a single revision: "over North China" has been replaced by "at a coastal background site in North China", so the title now states the single-site scope explicitly. The subtitle was adjusted at the same time to "nitrate amplification during high-PM_{2.5} conditions", which confines the nitrate claim to the polluted subset in which it is observed.

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

Technical correction 2. Use "enrichment factor" or "enrichment ratio" consistently (both currently appear in Sect. 3.1 and Sect. 3.2) and state the calculation once.

Response. Agreed. This inconsistency, also noted by Referee #1, has been removed: the term "enrichment ratio" is now used throughout the manuscript, the Supplement, and the figure axes and captions. The calculation is stated once, in Sect. 3.1, and the single definition covers both of the quantities to which the term is applied: for chemical components, the mass fraction in stage-relative high-PM_{2.5} samples divided by the corresponding screened stage-average fraction, and for PMF factors, the relative factor contribution in the high-value subset divided by its screened stage-average contribution. The Fig. 3b axis label and caption were updated to match.

Revised manuscript text (Sect. 3.1; p. 8, lines 260-265): For chemical components, the enrichment ratio is the mass fraction in stage-relative high-PM_{2.5} samples divided by the corresponding screened stage-average fraction; for PMF factors, it is the relative factor contribution in the high-value subset divided by its screened stage-average contribution. Ratios were calculated from unrounded values.

Revised manuscript text (Fig. 3 caption; p. 8, lines 275-280): (b) Enrichment ratios relative to screened stage-average fractions; the dashed line marks a ratio of 1.

Technical correction 3. Define all error bars and statistical summaries directly in the relevant figure captions.

Response. Agreed. This request, which Referee #1 also made, has been implemented for every figure: each caption in the main text and the Supplement now defines its statistical summaries and error bars directly, or states explicitly that no uncertainty intervals are shown. Specifically, the Fig. 1 caption defines the violin widths, boxes, medians, whiskers, and the bootstrap 95 % confidence intervals on stage means; the Fig. 2 caption defines the distribution elements and the stage-specific concentration cutoffs for selecting high-PM_{2.5} samples; the Fig. 3 caption specifies mean sample-level mass fractions, ratios of mean mass fractions, and ratios of group-mean ion concentrations, with no error bars; the Fig. 4 caption states that values are group-mean relative contributions reported to one decimal place with no error bars; the Fig. 5 caption defines the medians and interquartile bars; and the Fig. S9 caption states the frequency basis and that no uncertainty intervals are shown.

Revised manuscript text (Fig. 1 caption; p. 6, lines 210-215): Figure 1. Stage-level changes in PM_{2.5} and key chemical components at Tuoji Island during 2012–2023. (a) PM_{2.5} distributions; violin widths show distributions, boxes show interquartile ranges, centre lines show medians, whiskers extend to 1.5 times the interquartile range, grey points show a subsample of observations for visualisation, and coloured points and labels show means. (b, c) Bars show stage means, and error bars show bootstrap 95 % confidence intervals for the means. (d) Ratios were calculated from stage-mean concentrations. Shading denotes the three stages, and n is the number of valid samples.

Revised manuscript text (Fig. 2 caption; p. 7, lines 255-260): Figure 2. Identification and sensitivity tests for stage-relative high-PM_{2.5} samples after removal of mineral-dust outliers. (a) PM_{2.5} distributions after screening; violin widths show distributions, boxes show interquartile ranges, centre lines show medians, and whiskers extend to 1.5 times the interquartile range. Red dashed segments mark the stage-specific concentration cutoffs for selecting high-PM_{2.5} samples, and labels give the high-value and screened sample counts. (b) Bars show mean PM_{2.5} concentrations under the highest 10 %, 20 %, and 30 % definitions; labels give means and n. The horizontal dashed line is the fixed 75 µg m⁻³ reference.

Revised manuscript text (Fig. 3 caption; p. 8, lines 275-280): Panel (a) shows means of sample-level mass fractions, panel (b) shows ratios of these mean fractions, and panel (c) shows ratios of group-mean NO₃⁻ and SO₄²⁻ concentrations. No error bars are shown.

Revised manuscript text (Fig. 4 caption; p. 10, lines 320-325): Values are group-mean relative contributions and are reported to one decimal place; no error bars are shown.

Revised manuscript text (Fig. 5 caption; p. 12, lines 380-390): Figure 5. Stage changes in particle-phase ion balance and model-diagnosed aerosol liquid water content for all samples and stage-relative high-PM_{2.5} samples included in the ISORROPIA-II calculations. (a) NH₄⁺/2SO₄²⁻ molar ratio; (b) particulate NH₄⁺ excess after sulfate neutralisation; (c) metastable-mode ALWC. Open points denote all samples and filled points denote the high-value subset. Points and labels show medians, and horizontal bars show interquartile ranges.

Revised Supplement text (Fig. S9; p. 18): Frequencies are based on individual trajectories rather than daily modal assignments; equal-season-standardised stage frequencies are reported in Text S6. No uncertainty intervals are shown.

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