

Cross-phase partitioning of sulfur-nitrogen ratios and aerosol mixing-state evolution based on single-particle observations

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Abstract

The balance between sulfur and nitrogen (S-N) precursors influences secondary aerosol formation, yet its cross-phase links to particle composition and mixing states remain poorly constrained. Here, we integrated single-particle aerosol mass spectrometry (SPA-MS) with pollutant and meteorological observations during winter and summer emission control periods (ECPs) and corresponding normal periods (NPs) in Yangzhou, eastern China. By establishing gas-phase (gSNR), particle-phase (pSNR), and number-based (nSNR) sulfur-to-nitrogen ratio metrics, we combined causal inference with interpretable machine learning to analysis the underlying drivers. During ECPs, reduced NO_x emissions alongside relatively stable SO_2 concentrations elevated gSNR, driving an average $\sim 57\%$ increase in pSNR across particle classes and a 27% expansion in mean vacuum aerodynamic diameter (D_{va}) relative to NPs. Causal analysis revealed a stepwise propagation of S-N partitioning running sequentially from gas-phase precursors to single-particle chemistry and ultimately to the population mixing state. Particle-resolved chemistry demonstrated that pSNR enrichment was highest in particles containing both black carbon and organic carbon, but markedly lower in BC-free inorganic particles, indicating strong particle-type-dependent aging pathways. Relative humidity (RH) acted as a primary regulator of this cross-phase coupling: below 55% RH, the SNR metrics decoupled, maintaining substantial chemical heterogeneity across particle classes (externally mixed state); above 85% RH, aerosol liquid water uptake accelerated gas-particle exchange and multiphase processing, causing the SNR metrics to converge as aerosols evolved toward an internally mixed state. These findings suggest that future air-quality models would benefit from incorporating particle-type heterogeneity and humidity-dependent multiphase processes under shifting emission regimes.

1. Introduction

Secondary aerosols, dominated by sulfate and nitrate, constitute a major fraction of fine particulate matter ($\text{PM}_{2.5}$, aerodynamic diameter $\leq 2.5 \mu\text{m}$) (Huang et al., 2014; Wang et al., 2013). These species strongly influence aerosol hygroscopicity, toxicity, optical properties, and radiative forcing (Nozière et al., 2010; Sheldon et al., 2023; El Haddad et al., 2024). They originate mainly from the multiphase oxidation of sulfur dioxide (SO_2) and nitrogen oxides ($\text{NO}_x = \text{NO}_2 + \text{NO}$) (Cheng et al., 2016; Liu et al., 2022) and their formation is governed by the interplay of precursor abundance, atmospheric oxidizing capacity, and meteorological conditions (George and Abbatt, 2010; Shiraiwa et al., 2011; Wen et al., 2018). Among these factors, relative humidity (RH) plays a central role by regulating aerosol liquid water and phase state, which in turn influences surface properties, mass-transport rates, and chemical aging pathways (Shiraiwa et al., 2011; Liu et al., 2018; Huang et al., 2023). These coupled processes are concisely reflected in the sulfur-to-nitrogen ratio (SNR), a common diagnostic for source attribution and sulfate-nitrate (S-N) evolution (Xie et al., 2020; Liu et al., 2020; Sosa Echeverría et al., 2023). However, SNR is still often interpreted using bulk or time-averaged measurements (Snider et al., 2016; Sun et al., 2016; Xu et al., 2019), which may inherently obscure particle-type heterogeneity and mixing-state diversity that matter for multiphase processing. Furthermore, current model parameterizations often oversimplify the effects of RH and aerosol phase state (Schmedding et al., 2020; Shiraiwa et al., 2017), while observational constraints remain largely limited to heavily polluted episodes (Huang et al., 2023; J. Wang et al., 2021). These limitations leave uncertainties regarding how shifts in gas-phase precursor composition translate into particle composition, number fractions, and transitions between internal and external mixing, particularly under cleaner urban conditions.

These uncertainties may be magnified by the rapid evolution of anthropogenic emissions. Between 2013 and 2024, China's clean air actions triggered an asymmetric decline in major gaseous precursors (Y. Zhang et al., 2020; Geng et al., 2024): annual SO_2 fell from 40 to $8 \mu\text{g m}^{-3}$ (−80%) and stabilized well below the World Health Organization (WHO) guidelines, while NO_2 reductions were more moderate (−55%), remaining nearly twice the revised WHO annual limit (Ministry of Ecology and Environment of the People's Republic of China. Bulletins on the State of China's Ecological and Environmental Quality, 2013–2024. <https://www.mee.gov.cn/hjzl/sthjzk/zghjzkgb/>). This imbalance drove the national gas-phase SNR (gSNR) down from 1.27 to 0.56 and favored nitrate-dominated aerosols (Liu et al., 2022; Chu et al., 2020; Lin et al., 2023). However, further trend shifts are likely as policy priorities move toward deeper NO_x control. The WHO tightened its annual NO_2 guideline (from 40 to $10 \mu\text{g m}^{-3}$), while relaxing the daily SO_2 limit (from 20 to $40 \mu\text{g m}^{-3}$). China has strengthened industrial and traffic NO_x emission standards (Lu et al., 2020; Y. Zhang et al., 2021), and accelerated the uptake of new-energy

vehicles (Liang et al., 2019). Under this trend, gSNR may rise again, potentially coinciding with higher ozone (O_3) levels and a more oxidizing atmosphere (Li et al., 2019). The COVID-19 lockdowns provide a natural experiment consistent with this direction. Across Chinese megacities, lockdown periods induced sharp, short-term NO_x declines (30% to 50%) and O_3 increases (21% to 77%) while SO_2 remained stable (Le et al., 2020; Huang et al., 2021; Dai et al., 2024), effectively simulating future emission trends within a cleaner and more oxidizing background.

In this study, we use these perturbations in Yangzhou, a representative city in the Yangtze River Delta (YRD), to investigate multiphase S-N evolution across two lockdown-recovery cycles. By integrating single-particle aerosol mass spectrometry (SPA-MS) with causal inference and interpretable machine-learning analysis, we track S-N partitioning across gas, particle and number domains. This particle-resolved approach links precursor shifts, oxidizing conditions, and RH-dependent phase transitions to single-particle aging and mixing-state evolution, providing process-level evidence to improve regional model representations of coupled S-N partitioning under ongoing NO_x mitigation.

2. Methods

2.1 Site Description and Sampling Periods

Field observations were conducted at a national air-quality monitoring station in Yangzhou (32.41°N, 119.40°E), a representative urban center within the YRD of China. The monitoring site is situated 20 m above ground level in a mixed residential, commercial, and educational district, representing typical urban background conditions in the region. To examine atmospheric responses to abrupt emission changes during the COVID-19 pandemic, we selected four rain-free five-day episodes with broadly comparable meteorological conditions, comprising two emission control periods (ECPs) and their corresponding normal periods (NPs): a winter ECP (W-ECP, 29 January–2 February 2020) and its winter NP (W-NP, 20–24 February 2020), as well as a summer ECP (S-ECP, 17–21 August 2021) and its summer NP (S-NP, 17–21 September 2021).

Single-particle chemical composition, vacuum aerodynamic diameter (D_{va}), and mixing state were measured in real time using a single-particle aerosol mass spectrometer (SPA-MS; Hexin Analytical Instrument Co., Ltd., China). The instrument primarily detected particles with D_{va} ranging from 0.2 to 2.0 μm , as detailed previously (Dai et al., 2024). Concurrently, $PM_{2.5}$ mass concentrations were measured by a particulate matter monitor (XHPM2000E, Xianhe, China), while trace gases (NO_x , SO_2 , CO, and O_3) were quantified using Thermo Scientific analyzers (Models 42i, 43i, 48i, and 49i) (Figure S1). All instruments were co-located to ensure the temporal and spatial consistency of the aerosol and precursor datasets.

Meteorological parameters were retrieved via Google Earth Engine (GEE) from the NASA Goddard Earth Observing System Composition Forecast system (GEOS-CF) and the ERA5-Land hourly dataset from the European Centre for Medium-Range Weather Forecasts (ECMWF). Ten variables were extracted for the grid cell nearest to the monitoring site (**Table S2**): 2 m air temperature (T2m), 10 m air temperature (T10m), eastward and northward wind components at 2 m (U2m, V2m), surface net solar radiation (SSR), planetary boundary layer height (BLH), sea level pressure (SLP), total precipitation (TPREC), 2 m dew point temperature (DP2m), and relative humidity (RH). These variables were selected for their potential roles in influencing atmospheric stability, pollutant transport, and chemical transformation kinetics. All datasets were temporally synchronized to an hourly resolution to facilitate integrated analysis with the chemical measurements.

2.2 Single-Particle Classification

A total of 324,090 ambient particles were detected and classified based on their SPAMS mass spectra. Particles were clustered using the ART-2a algorithm in MATLAB R2024a (MathWorks, Inc.) via the Computational Continuation Core toolkit (COCO v1.4). The algorithm was executed for 20 iterations with a vigilance factor of 0.8 and a learning rate of 0.05, parameters consistent with established single-particle aerosol studies (Huang et al., 2023; G. Zhang et al., 2021; Dai et al., 2024). Following the initial clustering, particle types were aggregated into five operational principal groups according to their dominant mass-spectral signatures. Black Carbon (BC) particles were identified by carbon-cluster ions (e.g., m/z $\pm 12[C]^{+/-}$, $\pm 36[C_3]^{+/-}$, $\pm 48[C_4]^{+/-}$, and $\pm 60[C_5]^{+/-}$). BC with organic carbon (BCOC) particles exhibited the same BC signatures together with characteristic organic-carbon (OC) fragments (e.g., m/z $27[C_2H_3]^+$, $29[C_2H_5]^+$, $37[C_3H]^+$, and $43[C_2H_3O]^+$). BC-free inorganic (BF) particles lacked BC cluster signatures and were dominated by inorganic species. BC-free OC-rich (BFOC) particles also lacked BC clusters but showed a high relative abundance of OC fragments. Residual subclasses with mixed or ambiguous signatures were defined as “Other”.

To better resolve S-N partitioning, we further divided these principal groups into 19 subclasses. Subclasses enriched in secondary inorganic species (SIS) were identified by sulfate ($-97[HSO_4^-]$, $-96[SO_4^-]$), nitrate ($-62[NO_3^-]$, $-46[NO_2^-]$), or both, and were labeled with the suffixes “-S”, “-N” and “-SN”, respectively. Particles exhibiting strong cyanide-related ions ($-26[CN^-]$ and $-42[CNO^-]$) were labeled with “-CN”. Number fractions and representative mass spectra are summarized in Supplementary **Table S1** and **Figure S2**.

2.3 Classification of Fresh-like and Aged-like Particles

Aerosols were further grouped into fresh-like (-CN) and aged-like (-sec) based on diagnostic SPA-MS fragments and D_{va} (**Figure 1f** and **Figure 2**). Strong cyanide-related ions together with carbonaceous fragments are commonly observed in relatively fresh combustion particles from biomass burning, coal combustion, and vehicle exhaust (Huang et al., 2023; Sun et al., 2017; G. Zhang et al., 2020). We therefore treated CN-containing subclasses within each principal group as operational fresh-like states. For instance, BCOC-CN particles exhibit abundant carbon-cluster and hydrocarbon fragments accompanied by cyanide signatures (**Figure 2h**), occurring predominantly at smaller D_{va} (100-600 nm), consistent with freshly emitted combustion aerosols (Sharma et al., 2018). Similarly, BFOC-CN particles display $+39[K^+]$, $-26[CN^-]$, and levoglucosan-derived fragments ($-45[CHO_2^-]$, $-59[C_2H_3O_2^-]$, $-73[C_3H_5O_2^-]$) and are predominantly found at small sizes, representing primary organic aerosol from biomass burning (**Figure 2c**) (Bi et al., 2011; Huang et al., 2023).

Aged-like types (-sec) were constructed by regrouping sulfate-rich (-S), nitrate-rich (-N) and mixed (-SN) subclasses into composite SIS-enriched categories within each principal group, reflecting extensive secondary processing (Xie et al., 2020). BCOC-sec types display coexisting BC, OC and strong SIS fragments (**Figure 2g**), indicating substantial internal mixing of carbonaceous and secondary inorganics, in line with previous observations (Dai et al., 2024; Xie et al., 2020). BC-sec (BC-S, BC-N, BC-SN; **Figure 2f** showed negligible OC signals but elevated SIS ions, while BF-sec and BFOC-sec (BF-S, BC-N, BC-SN; BFOC-S, BC-N, BC -SN) exhibit elevated sulfate and nitrate ions but lack BC fragments (**Figure 2b, d**).

The consistency of this fresh-aged framework is supported by both compositional and temporal variations. The relative contribution of SIS signal is systematically lower in “-CN” than in “-sec” types for BC, BCOC, BF and BFOC groups (**Figure S3a**). In addition, hourly number concentrations of each “-sec” type correlate most strongly with the “-CN” type within the same principal group (**Figure S3b**). Together with the systematic increase in D_{va} from “-CN” to “-sec”, these diagnostics support interpreting “-CN” and “-sec” as fresh-like and aged-like compositional states within each principal group. We therefore define four dominant fresh-like/aged-like pairs: BC-CN/BC-sec, BCOC-CN/BCOC-sec, BF-CN/BF-sec, and BFOC-CN/BFOC-sec.

2.4 Definition of Sulfur-to-Nitrogen Ratios Metrics

To quantify S-N partitioning across gas, particles, and number domains, we define three complementary SNR metrics: gas-phase SNR (gSNR), particle-phase SNR (pSNR), and number-based SNR (nSNR). The gSNR is calculated as the molar ratio of SO_2 to NO_x from hourly observations:

$$\text{gSNR} = \frac{[\text{SO}_2]}{[\text{NO}_x]}.$$

207

208 The pSNR is derived from SPA-MS spectra by calculating the ratio of total peak areas
 209 (PA) of sulfate ions ($-97[\text{HSO}_4^-]$, $-96[\text{SO}_4^-]$) to nitrate ions ($-62[\text{NO}_3^-]$, $-46[\text{NO}_2^-]$)
 210 for a selected particle or particle class:

$$\text{pSNR} = \frac{\text{PA}_{\text{sulfate}}}{\text{PA}_{\text{nitrate}}}.$$

212

213 Although SPA-MS is semi-quantitative, uniform acquisition and processing of sulfate-
 214 and nitrate-related ion signals provide a consistent basis for comparing relative
 215 variations in pSNR. Because ion responses and matrix effects may vary, pSNR is
 216 interpreted as a relative spectral indicator rather than an absolute sulfate-to-nitrate
 217 molar or mass ratio, consistent with prior SPA-MS studies (Healy et al., 2013; G. Zhang
 218 et al., 2021).

219

220 The nSNR represents the ratio of the number concentrations (N) of sulfur-containing
 221 particles to nitrogen-containing particles, serving as a population-level proxy for
 222 external mixing tendency of sulfate- versus nitrate-containing particles:

$$\text{nSNR} = \frac{N_{\text{sulfur-contai}}}{N_{\text{nitrogen-con}}}.$$

224

225 2.5 Particle-Type-Resolved Hygroscopicity Estimation

226 To characterize particle-type-resolved hygroscopicity, we inferred an effective
 227 hygroscopicity parameter (κ_{eff}) for each particle class at hourly resolution using
 228 aggregated SPA-MS ion signals. Major ion peaks were assigned to several chemical
 229 components (e.g., sulfate, nitrate, organics, black carbon, and potassium salts). Sulfate
 230 and nitrate were represented by ammonium sulfate and ammonium nitrate, respectively.
 231 For each component i , the hourly summed peak area A_i was normalized by its assumed
 232 density ρ_i (e.g., ammonium nitrate for nitrate, ammonium sulfate for sulfate; **Table S3**)
 233 to obtain an equivalent volume. The volume fraction of component i was then
 234 approximated as

$$\varepsilon_i \approx \frac{A_i/\rho_i}{\sum_j (A_j/\rho_j)}.$$

236

237 The effective hygroscopicity of each particle class was calculated using κ -Köhler theory
 238 and the Zdanovskii-Stokes-Robinson (ZSR) mixing rule (Petters and Kreidenweis,
 239 2007):

$$\kappa_{\text{eff}} = \sum_i (\varepsilon_i \times \kappa_i),$$

241 where κ_i is the literature-derived hygroscopicity parameter of component i . **Table S3**
 242 lists the lower, mean, and upper κ_i values for ammonium sulfate and ammonium nitrate

and the prescribed values for the other components. The mean values were used in the baseline calculation. Sensitivity was evaluated using four combinations of the lower and upper values for ammonium sulfate and ammonium nitrate (lower-lower, lower-upper, upper-lower, and upper-upper), while the component volume fractions, densities, κ_i values of the other components, and ZSR mixing rule were held constant.

The resulting κ_{eff} represents an hourly, particle-type-resolved estimate derived from aggregated SPA-MS spectra. SPA-MS ion signals are semi-quantitative and may be affected by species-dependent ionization efficiencies and matrix effects. The estimates also assume ammonium salts for sulfate and nitrate and volume-additive mixing under the ZSR rule. Accordingly, κ_{eff} is interpreted as a relative indicator for comparisons among particle classes, periods, and aging states, rather than as a direct hygroscopicity measurement.

2.6 Causal-Structure Analysis

Causal networks, expressed as directed acyclic graphs (DAGs), characterize conditional dependence structures and identify directed relationships compatible with potential causal pathways among variables (Koller, 2009). In this study, we applied the Causal Learner toolbox in MATLAB R2024a (MathWorks, Inc.) (Ling et al., 2022) to explore potential relationships among SNR metrics, meteorological parameters, and gaseous pollutants.

For each target SNR metric (gSNR, pSNR, nSNR), the Grow-Shrink (GS) algorithm was first applied to determine its Markov blanket (MB), the minimal variable set directly linked to the target. The GS procedure alternates between adding conditionally dependent variables (grow phase) and removing conditionally independent variables (shrink phase) until convergence (Margaritis, 1999). The MB-derived subset was further refined using the Greedy Equivalence Search (GES) algorithm, which iteratively improves the global causal structure via adding edges (forward phase) and removing redundant links to prevent overfitting (backward phase) (Chickering, 2003). The resulting DAGs identified direct and indirect dependency pathways linking SNR evolution to meteorological and chemical drivers. The statistical support and physical plausibility of these pathways provide a process-consistent structural basis for subsequent predictive modeling.

2.7 SSA-XGBoost Modeling and Interpretation

Building on the DAG-derived causal structure, we used the eXtreme Gradient Boosting (XGBoost) algorithm to construct separate models for each aged-like particle class (BCOC-sec, BC-sec, BFOC-sec, BF-sec), with hourly pSNR and nSNR modeled in turn as target variables, to quantify the relative influence of the identified drivers on

their variability. All predictors were standardized by the z-score method before model fitting. The dataset was split into six folds: five folds for iterative training and validation, and one held-out fold reserved exclusively for interpretation to prevent information leakage. Model hyperparameters were tuned via the Sparrow Search Algorithm (SSA), a swarm-intelligence metaheuristic that balances global exploration (discoverer phase) and local exploitation (joiner phase) to prevent premature convergence (Yang et al., 2025). SSA was run with a population size of 10 and 50 iterations. Model performance was assessed using root-mean-square error (RMSE) and the coefficient of determination (R^2), averaged over five validation folds (**Table S4**). The SSA-XGBoost model achieved high R^2 (0.71–0.91) for major particle types, ensuring the robustness of the predicted SNR values.

We interpreted the trained models using SHapley Additive exPlanations (SHAP), a cooperative game-theoretic approach that decomposes individual predictions into additive feature contributions (Ekanayake et al., 2022). SHAP values quantify both the magnitude and direction of each predictor's effect on pSNR or nSNR. This approach provides transparent and physically meaningful insights into how gSNR, meteorology, and gaseous pollutants modulate SNR variability across aged-like particle types.

3. Results and discussion

3.1 Atmospheric Responses to Emission Shifts

Using the four episodes defined in **Section 2.1**, we assessed atmospheric responses to abrupt anthropogenic emission shifts across two COVID-19 ECP and NP cycles in Yangzhou (**Figure 1** and **Figure S1**). During the ECPs, surface NO_x concentrations decreased sharply relative to the corresponding NPs, by 39% in winter and 34% in summer (**Figure 1a**). In contrast, SO_2 changed little and remained within 3.5 to 3.9 ppb. This asymmetric response significantly enhanced gSNR, with seasonal averages increasing from 0.21 to 0.36 (+71%) in winter and from 0.35 to 0.56 (+60%) in summer. O_3 was about 15% higher during ECPs, consistent with weaker NO titration (Wang et al., 2022). $\text{PM}_{2.5}$ decreased modestly during ECPs, by 13% in winter and 9% in summer (**Figure 1a**).

Time series show that gSNR, pSNR, and nSNR varied synchronously, and their fluctuations were more pronounced during ECPs than during NPs (**Figure 1c**). This co-variation indicates that changes in precursor composition were accompanied by coherent responses in particle composition and in the population-level abundance of sulfur- and nitrogen-containing aerosols. Total particle number concentration (N) increased during NPs while $\text{PM}_{2.5}$ mass remained similar, suggesting a shift toward smaller particles that contribute more strongly to number than to mass after controls were relaxed (**Figure 1d**).

As shown in **Figure 1b**, BC-containing particles dominated the aerosol population across all episodes, accounting for an average of 58% of total detections and peaking at 69% during S-ECP. Freshly emitted particles accounted for approximately 16% of detections during ECPs and rose to ~26% in the NPs, reflecting the rebound in primary emissions. Relative to NPs, ECPs were associated with more sulfate-dominated secondary inorganic species (SIS) and pronounced particle growth across particle classes. Averaged across all categories, pSNR was about 57% higher during ECPs than during NPs (**Figure 1e**), with a stronger shift in BC-containing than in BC-free particles and a larger contrast in summer than in winter. Mean D_{va} was increased by about 27% during ECPs, with the largest increases in aged-like types (**Figure 1f**). Across all principal groups, aged-like categories consistently showed higher pSNR and broader size distributions than their fresh-like counterparts, consistent with enhanced oxidation under elevated gSNR, favoring sulfate-rich growth.

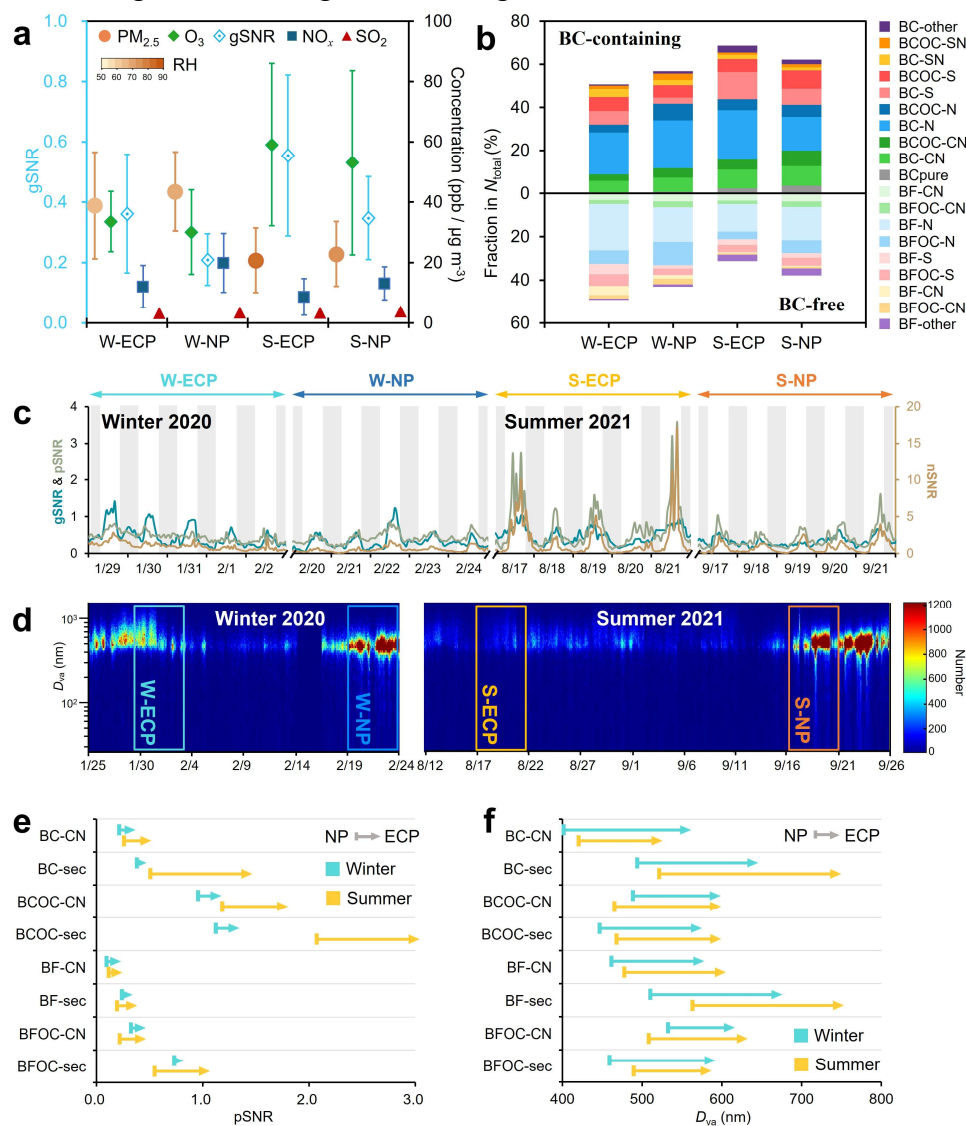


Figure 1. Physicochemical characteristics of particles under different emission control and normal periods. W-ECP, W-NP, S-ECP and S-NP denote the winter emission

HONO/NO₂ release (Liang et al., 2021; Ye et al., 2017) and photo-induced formation of reactive oxygen species (ROS) that accelerate the conversion of SO₂ to sulfate (Zhang et al., 2022; Zhu et al., 2020). At night, when solar radiation is negligible and RH is high (>80%), λ_{OC} exceeds λ_{BC} , aligning with aqueous-phase sulfate production through H₂O₂ or O₃ oxidation and with possible organosulfate formation (Cheng et al., 2016; Y. Wang et al., 2021). Increased organic-phase viscosity may further limit gas-particle partitioning (Vaden et al., 2011), water uptake (Rickards et al., 2015), and multiphase reactions (Shiraiwa et al., 2011), which could prolong sulfate retention within OC-rich particles. Laboratory and modeling studies also suggest that BC can catalyze SO₂ oxidation even in darkness (He et al., 2018; He and He, 2020), although this likely represents a minor nighttime pathway compared with OC-driven aqueous processes.

Across the summer diurnal cycle, λ_{BCOC} consistently exceeds both λ_{BC} and λ_{OC} , peaking in the afternoon (**Figure 3d**). This persistent enhancement suggests a synergistic role of BCOC, BC provides reactive or catalytic surfaces, while OC facilitates sulfate accommodation or retention within the particle matrix. Such synergy likely reflects the complex surface chemistry of BC, involving polycyclic aromatic hydrocarbons (PAHs) and aromatic oxygenated species, which may promote SO₂ uptake and facilitate heterogeneous oxidation under ultraviolet (UV) irradiation (Riva et al., 2016). Light-absorbing organic compounds, such as brown carbon and humic-like substances, may further act as photosensitizers and accelerate SO₂-to-sulfate conversion (X. Wang et al., 2020).

In winter (**Figure 3a, b**), all λ values are smaller, but the compositional contrasts remain. λ_{OC} generally exceeds λ_{BC} throughout the day, consistent with a larger relative contribution of OC-related aqueous pathways under weaker photochemistry. Nevertheless, λ_{BCOC} remains the largest, indicating that BCOC mixtures still provide the most favorable environment for sulfate enrichment even under low-radiation, low-temperature conditions. This persistent BCOC advantage may reflect not only enhanced photochemical/heterogeneous processing, but also microphysical effects, such as larger accessible surface associated with soot morphology (Beeler et al., 2025) and coating softening or water uptake (Li et al., 2021) that promotes sulfate partitioning and retention.

These observations show that the diurnal variation of pSNR is strongly composition-dependent. BC-containing particles show the strongest enhancement during photochemically active hours, OC-rich particles become relatively more important under humid nighttime and winter conditions, and mixed BCOC particles show the largest enhancement in both seasons, likely driven by combined chemical and microphysical effects.

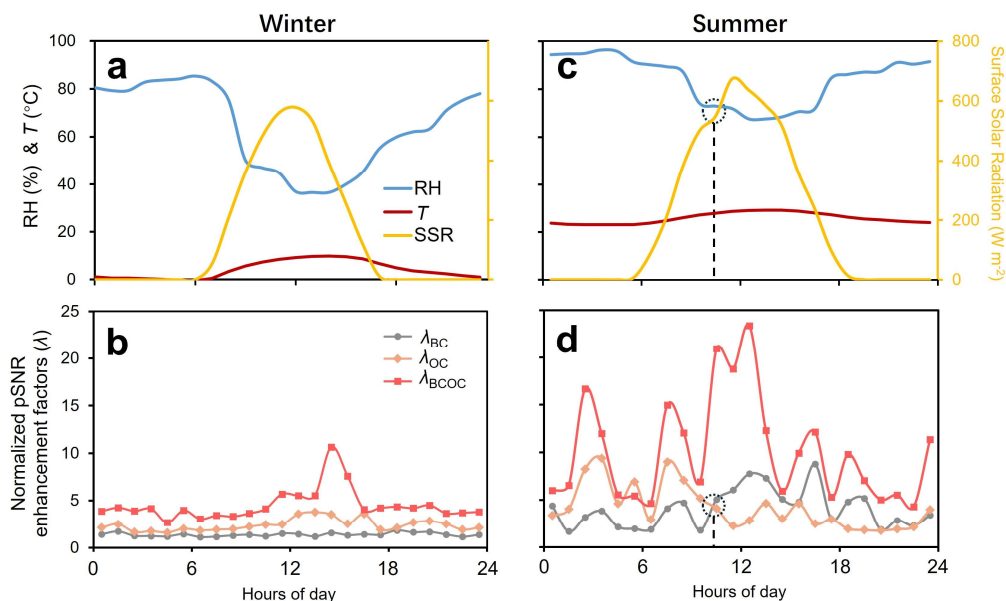


Figure 3. Diurnal cycles of meteorology and component-resolved particle-phase sulfur-to-nitrogen ratio (pSNR) enhancement (λ) in winter (a, b) and summer (c, d). (a, c) show the diurnal variation of meteorological parameters, including relative humidity (RH, blue), temperature (T , deep red), and surface solar radiation (SSR, yellow). (b, d) present normalized pSNR enhancement factors (λ) values for components: organic carbon (λ_{OC} ; pink), black carbon (λ_{BC} ; black), and the mixed BC-OC (λ_{BCOC} ; red). The dashed circles indicate the transitional period when BC overtakes OC in driving pSNR enhancement during summer.

3.3 Particle-Type-Dependent Aging Patterns

To characterize particle-type-dependent aging, we jointly examined changes in inferred κ_{eff} , pSNR, and D_{va} and developed a conceptual framework in the κ_{eff} -pSNR space (Figure 4). As defined in Section 2.3, each fresh-like subtype was paired with its dominant aged-like counterpart. Stage I compares fresh-like and aged-like populations during NPs, whereas Stage II compares aged-like populations between NPs and ECPs. These comparisons represent population-level contrasts rather than trajectories of individual particles. To evaluate the robustness of inferred κ_{eff} , calculations were repeated using alternative hygroscopicity values for ammonium sulfate and ammonium nitrate (Table S3; Figure S4). The resulting ranges represent parameter sensitivity within the adopted mixing rule rather than the full uncertainty in κ_{eff} . Because relative ionization efficiencies and matrix effects can affect SPA-MS signals, pSNR changes are interpreted as relative ion-signal shifts rather than absolute composition changes.

For OC-rich fresh particles (BCOC-CN and BFOC-CN), Stage I aged-like populations had an approximately 5% lower mean D_{va} and an 83% higher pSNR than their fresh-like counterparts, whereas κ_{eff} changes were generally small or parameter dependent.

No consistent κ_{eff} increase or decrease was obtained for BCOC in either season or BFOC in winter, while the summer BFOC response exhibited (2.5%–17.0%). This pattern may reflect compaction and the formation of an organic-rich shell that retains sulfate while limiting additional water uptake (Mikhailov et al., 2009; Riemer et al., 2019). During Stage II, stronger oxidation is associated with an increase in D_{va} of about 25% and a further rise in pSNR of about 80%, again with minimal change in κ_{eff} , consistent with continued sulfate accumulation and coating growth on a compact and relatively hydrophobic matrix.

For BC particles with weaker organic signatures, the transition from BC-CN to BC-sec exhibited the largest κ_{eff} increase ($\sim 31\%$). This response remained positive across all tested parameter combinations, ranging from 16.8% to 27.5% in winter and from 28.5% to 47.1% in summer (**Figure S4**). Together with increases in D_{va} ($\sim 24\%$) and pSNR ($\sim 87\%$), this pattern indicates dominant inorganic uptake during NPs. During Stage II, pSNR increased markedly ($\sim 105\%$) and D_{va} increased by $\sim 36\%$ to ~ 700 nm, while the inferred κ_{eff} increase remained small across the tested parameter combinations (1.0%–4.6%). This shift indicates enhanced sulfate-rich coating growth as gSNR and oxidant levels rise.

For BC-free particles (BF-CN to BF-sec), pSNR remained relatively low and changed only slightly. Stage I κ_{eff} changes remained positive but small in both winter (1.3%–5.6%) and summer (1.3%–7.3%). The Stage II κ_{eff} response was also modest in winter (0.9%–7.3%) but stronger in summer (11.6%–18.2%). Together with the approximately 22% increase in D_{va} , these changes are consistent with increased secondary inorganic contributions, particularly during summer ECPs.

Overall, the coupling between inferred κ_{eff} and pSNR strengthened under higher gSNR and stronger oxidative conditions, although the response remained particle-type dependent. BC-containing particles showed pronounced sulfate enrichment, consistent with the accumulation of secondary sulfate coatings on soot particles. OC-rich particles also exhibited strong pSNR enhancement but comparatively limited changes in inferred hygroscopicity, whereas BC-free particles showed weaker compositional adjustment and more gradual particle growth. Thus, the two-stage framework captures distinct composition-dependent responses rather than a uniform aging pathway, reflecting differences in core type, coating chemistry, and mixing state.

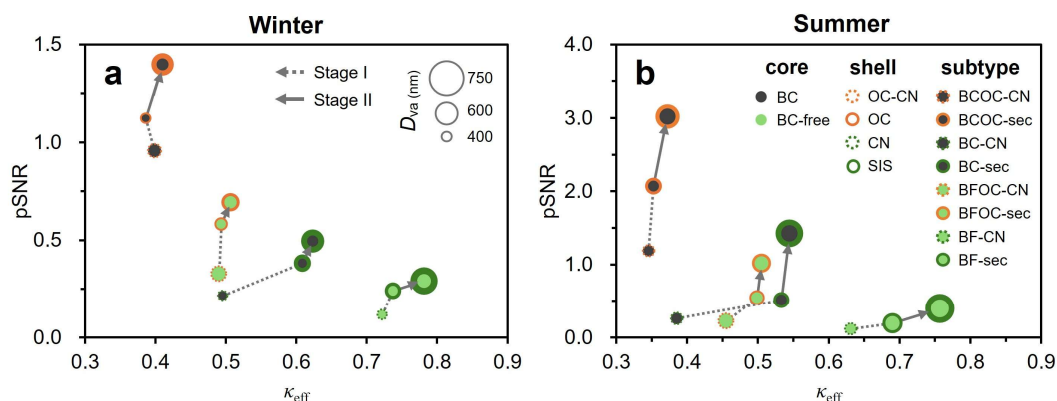


Figure 4. Conceptual framework for the co-evolution of particle-phase sulfur-to-nitrogen ratio (pSNR) and inferred hygroscopicity (κ_{eff}) among particle classes during winter (a) and summer (b). Particles are represented by core-shell schematics. The inner circle indicates BC-containing (dark gray) or BC-free (light green) particles, and the outer ring indicates coatings dominated by organic carbon (OC, orange) or secondary inorganic species (SIS, green). Dashed and solid rings denote fresh-like and aged-like populations, respectively. Marker size scales with vacuum aerodynamic diameter (D_{va}). Shell thickness estimates from the D_{va} of aged-like and fresh-like particles. Dotted and solid arrows indicate aging relationships during Stage I in normal periods (NPs) and additional Stage II processing in emission control periods (ECPs), respectively.

3.4 Humidity-Dependent Sulfur-Nitrogen Partitioning

Ammonium sulfate (AS) and ammonium nitrate (AN) are the dominant SIS in $\text{PM}_{2.5}$ (Fu et al., 2016; Lin et al., 2020). Their deliquescence relative humidity (DRH), together with the mutual deliquescence RH (MDRH) of their mixtures, provide reference points for interpreting particle phase transitions and aqueous processing (Liu et al., 2021; Sun et al., 2018). These transitions may shift in the presence of organic constituents and other components in ambient particles (Li et al., 2021). As RH increases, the associated rise in aerosol liquid water (ALW) reduce matrix viscosity, enhances molecular transport, and promotes morphological restructuring and internal mixing (Mikhailov et al., 2009; Shiraiwa et al., 2011). In our observations, increasing RH is accompanied by generally declining and progressively converging gSNR, pSNR, and nSNR, indicating a shift toward more balanced S-N partitioning across gas, particle, and number domains (Figure S5a, e). In parallel, BCOC-sec consistently exhibited the highest pSNR and nSNR, while BF-sec remains the lowest, but these contrasts weaken as RH increases, suggesting that elevated humidity reduces particle-type compositional and population-level contrasts of S-N partitioning (Figure S5b, f). Guided by these physicochemical thresholds and the observed SNR evolution, we divide the observations into four characteristic RH regimes (Figure 5). Because particle phase state and ALW were not directly measured, these regimes are operational ranges rather than fixed phase-transition boundaries.

Low-RH regime (RH < 55%). Under dry conditions, particles are largely solid with negligible ALW and limited ionic mobility (Sun et al., 2018). Uptake of HNO₃ and N₂O₅ hydrolysis is suppressed, restricting particulate nitrate formation (Bertram and Thornton, 2009), while SO₂ oxidation on BC and mineral surfaces proceeds slowly (He et al., 2018; Zhao et al., 2017). High solar radiation in this regime favors particulate nitrate photolysis, generating OH radicals that can drive heterogeneous SO₂ oxidation (He et al., 2017; Ye et al., 2017). As a result, pSNR is locally elevated, particularly in BCOC-sec, where BC serves as a photochemically active substrate enhancing sulfate formation, while OC acts as a stabilizing matrix that retains sulfate internally (**Section 3.2**). Weak correlations among gSNR, pSNR and nSNR (**Figure S6**) indicate kinetic limitations on gas-particle mass transfer and the particle population remains externally mixed with pronounced differences in nSNR across particle types (**Figure S5c, g**).

Moderate RH regime I (RH ≈ 55%–70%). Crossing the DRH of AN (~61%) induces a solid to semi-solid transition, forming a reactive aqueous surface film (Seinfeld and Pandis, 2016). These films relax interfacial diffusion constraints and enhance ionic mobility, activating solid particles by rapid water uptake, which is consistent with increases in particle size and number concentration (**Figure S5d, h**). ALW on BC surfaces may dissociate to OH radicals, while O₃, H₂O₂ and NO₂ oxidation pathways further enhance sulfate formation (He and He, 2020; J. Wang et al., 2020). Because NH₃ tends to neutralize H₂SO₄ preferentially, sulfate can remain relatively favored during the early stage of humidification, even as nitrate production increases (Wang et al., 2013; Seinfeld and Pandis, 2016). Accordingly, pSNR rises, most strongly in BC-containing classes such as BC-sec and BCOC-sec (**Figure S5b, f**).

Moderate RH regime II (RH ≈ 70%–85%). As RH approaches the MDRH of AS-AN mixtures (69%–75%), semi-solid particles transition toward fully liquid states with substantial ALW uptake and bulk-phase processing (Sun et al., 2018). This phase transition promotes efficient internal mixing and SIS shell formation (Riemer et al., 2019), driving particle growth and increased number concentration primarily through condensational uptake between ~60%–75% RH (**Figure S5d, h**). Once H₂SO₄ is largely neutralized, excess NH₃ facilitates NH₄NO₃ formation (Seinfeld and Pandis, 2016). High ALW and elevated effective Henry's law constants enhance HNO₃ uptake, while NO₂ hydrolysis and heterogeneous N₂O₅ reactions accelerate nitrate accumulation (Bertram and Thornton, 2009). These processes progressively shift S-N partitioning toward nitrogen, lowering pSNR from its peak near 70% RH. Concurrently, the differences among gSNR, pSNR, and nSNR narrow, suggesting that gas-particle S-N partitioning increasingly approaches quasi-equilibrium and cross-phase coupling strengthens (**Figure 5**). Around 80% RH, both D_{va} and particle number concentration show a sharp and transient decline, likely reflecting enhanced coagulation, temporarily reduced net condensational growth (El Haber et al., 2024), and liquid-coating-induced

restructuring (e.g., capillary compaction of loose aggregates into denser particles) (Chen et al., 2016; Beeler et al., 2025).

High-RH regime (RH > 85%). Exceeding the DRH of AS (~80 %) (Lightstone et al., 2000) induces a predominantly aqueous particle phase (Bateman et al., 2016; Sun et al., 2018). Under these conditions, dissolution of reactive precursors is favored and kinetic barriers to gas-to-particle partitioning are strongly reduced (Seinfeld and Pandis, 2016), leading to much tighter coupling among gSNR, pSNR, and nSNR (R^2 ranged from 0.68 to 0.81, **Figure S6**). All three SNR metrics converge to similarly low and relatively stable values (**Figure 5**), consistent with more efficient cross-phase exchange and multiphase processing. Particle-type SNR curves (**Figure S5b, c, f, g**) show minimal divergence, reflecting extensive internal mixing that homogenizes particle composition and convergence of external mixing states across the population. Near saturation, continued water uptake increases particle size and may enhance wet removal (e.g., fog/precipitation-related scavenging), contributing to reduced number concentrations (**Figure S5d, h**).

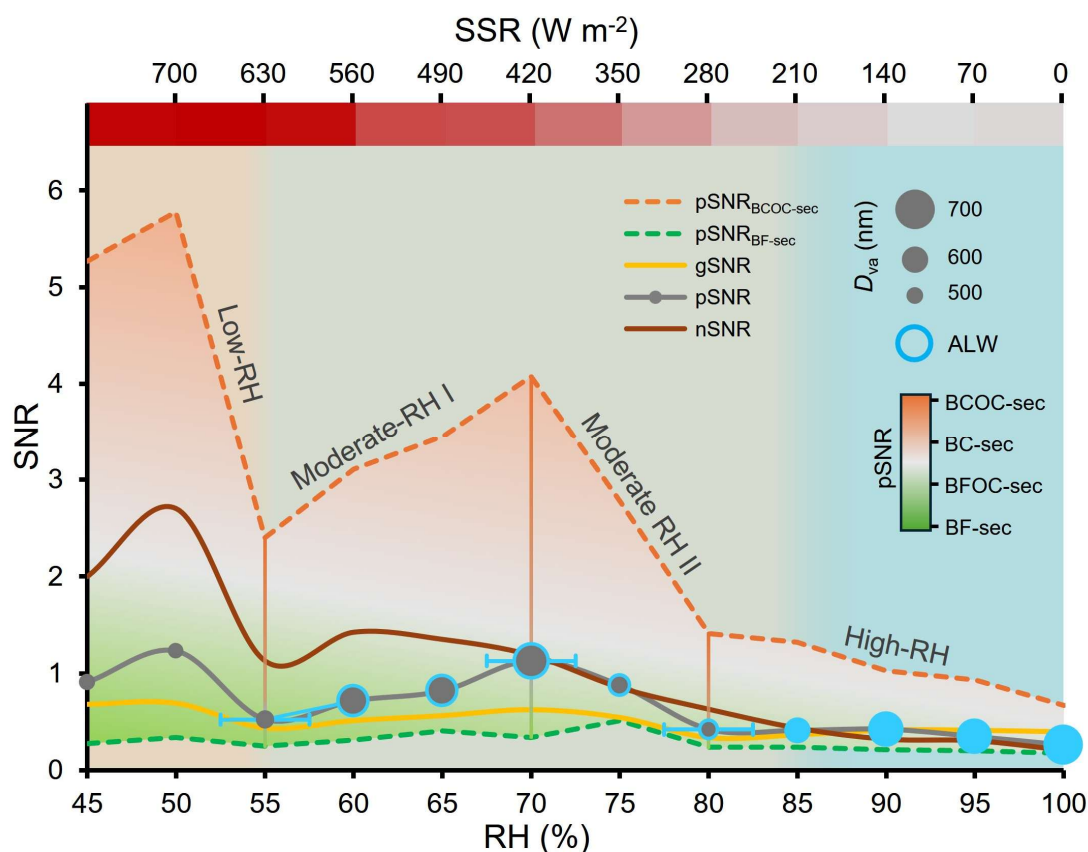


Figure 5 . Phase-state-resolved variations of sulfur-to-nitrogen ratios (SNRs) and particle physicochemical properties as a function of relative humidity (RH). Shown are RH-dependent variations of gas-phase (gSNR, yellow), particle-phase (pSNR, grey), and number-based (nSNR, brown), overlaid with pSNR of representative particle classes: BCOC-sec (orange dashed) and BF-sec (green dashed). Color shading beneath

the curves denotes the relative magnitude of pSNR, following the sequence BCOC-sec > BC-sec > BFOC-sec > BF-sec. The RH regimes are classified as Low RH (< 55%), Moderate RH (55%–70% and 70%–85%), and high RH (> 85%), with the top red gradient bar indicating mean surface solar radiation (SSR). Circular markers represent average particle size (bubble size: D_{va}), phase state (grey: solid; grey with blue ring: semi-solid; blue: liquid), and aerosol liquid water (ALW, blue ring). Vertical lines mark the DRH of NH_4NO_3 (AN), the MDRH of $(\text{NH}_4)_2\text{SO}_4$ - NH_4NO_3 mixtures (AN-SN), and the DRH of $(\text{NH}_4)_2\text{SO}_4$ (SN); error bars represent variability among literature reports and temperature-dependent shifts.

3.5 Causal Structure and Key Predictors of SNR Variability

Causal Network Insights. From the ten meteorological and chemical descriptors (Table S2), the GS algorithm selected eight key predictors: O_3 , T2m, SSR, RH, WS, WD, SLP, and BLH. Applying the GES to this subset produced the DAGs in Figure 6. In the DAGs, arrow direction denotes causal influence, arrow color encodes the sign of association (red for positive, grey for negative), and arrow thickness scales with the absolute Pearson correlation between connected variables. The DAG analysis identifies a consistent dependency structure linking gSNR to pSNR and nSNR. While these arrows indicate the most probable direction of influence under current statistical constraints, they represent the sequential propagation of chemical signals from gas-phase precursors to particle-level composition and then reshapes the population-level mixing state, consistent with the mechanistic picture developed in Sections 3.2-3.4, but may still be affected by unmeasured factors (e.g., NH_3 availability or heterogeneous kinetics) that are not explicitly represented in the network.

In addition, O_3 and BLH occupy intermediate positions between the SNR metrics and the meteorological cluster (RH, T2m, SSR, SLP), indicating that oxidant levels and boundary-layer dynamics may mediate the influence of radiation and stability on S-N partitioning. RH, T2m and SSR form a densely connected sub-network with predominantly negative links between RH, T2m and SSR, consistent with the anti-correlation between humidity and temperature/solar radiation. WS and WD show weaker and more diffuse connections, suggesting a secondary role through transport and dispersion. Overall, the DAG indicates that precursor ratios, oxidant levels and boundary-layer structure jointly organize the progression from gas-phase composition to particle-phase and number-based SNR metrics.

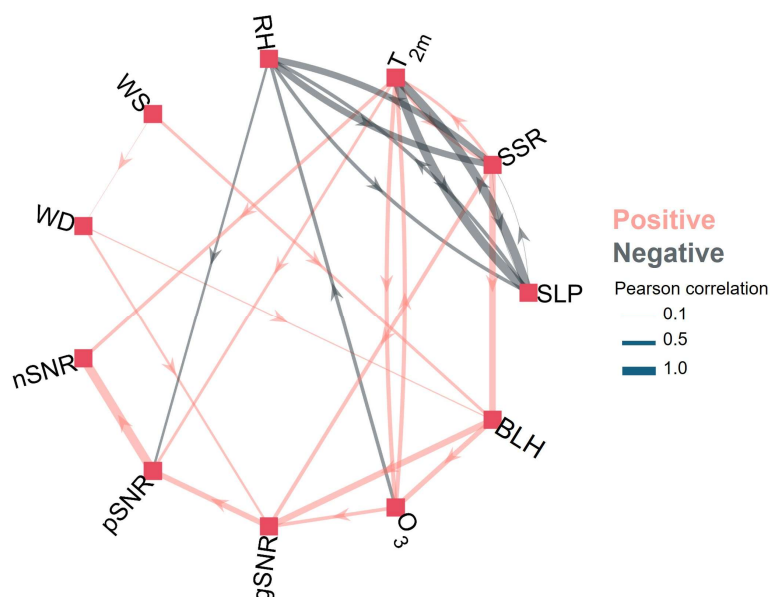


Figure 6. Causal network linking sulfur-to-nitrogen ratios (SNRs), meteorological variables, and ozone. Arrows indicate the directions of dependencies inferred from causal analysis. Red and gray arrows denote positive and negative Pearson correlations, respectively, with line thickness proportional to the absolute correlation coefficient. Variables include gas-phase (gSNR), particle-phase (pSNR), and number-based (nSNR) sulfur-to-nitrogen ratios; relative humidity (RH); air temperature at 2 m (T_{2m}); surface solar radiation (SSR); sea level pressure (SLP); boundary layer height (BLH); ozone (O₃); wind speed (WS); and wind direction (WD).

Model Performance Validation. To complement the causal analysis and capture nonlinear interactions, we trained particle-class-specific models using an SSA-optimized XGBoost framework. Cross-validated performance statistics (**Table S4**) show R^2 values above 0.70 with low RMSE for all three SNR metrics, indicating robust predictive skill across different particle types and limited overfitting.

SHAP-based Interpretation of Predictor Contributions. To quantify the relative influence of candidate drivers, we examined SHAP values for each SNR metric. As shown in **Figure S7a-d**, gSNR is the dominant determinant of particle-phase S-N partitioning, with the strongest influence for BC-sec, where large SHAP magnitudes suggest strong sensitivity of BC-containing particles to gas-phase composition. In the number domain (nSNR; **Figure S7e-h**), pSNR is the leading contributor across all aged-like classes, supporting a sequential response in which gas-phase variability first modulates within-particle S-N balance (pSNR) and is subsequently expressed at the population level (nSNR), consistent with the causal pathway inferred in **Section 3.5**.

Meteorological factors exert secondary, modulatory influences on SNR metrics. RH shows a consistent negative effect, reflecting enhanced nitrate formation and dilution

under humid conditions. Conversely, SSR and O₃ contribute positively (**Figure S7b, f**), indicating enhanced photochemistry activity and SO₂ oxidation (Seinfeld and Pandis, 2016; He et al., 2014). WD and BLH generally contribute negatively, capturing the dilution and vertical mixing effects that suppress SNR metrics (**Figure S7c, g**).

4. Conclusions and atmospheric implications

This study identified linked variations in gas-phase (gSNR), particle-phase (pSNR), and number-based (nSNR) sulfur-to-nitrogen ratios, connecting precursor composition with particle chemistry and population characteristics. During emission control periods (ECPs), NO_x decreased while SO₂ concentrations remained relatively stable, increasing gSNR. Compared with normal periods (NPs), pSNR was approximately 57% higher and mean vacuum aerodynamic diameter (D_{va}) was 27% larger. Causal analysis inferred directed links from gSNR to pSNR and from pSNR to nSNR, alongside connections with O₃ and meteorological variables. Predictor rankings from interpretable machine learning were consistent with this structure.

These relationships were accompanied by particle-type differences in diurnal composition patterns and population-level aging comparisons. Particles containing both black carbon (BC) and organic carbon (OC) showed the greatest pSNR enhancement relative to BC-free inorganic particles in both seasons. Among the other carbonaceous classes, enhancement was greater in BC particles with weaker organic signatures around midday in summer, but greater in BC-free OC-rich particles at night and in winter. The aging comparisons further showed that compositional changes were not uniformly accompanied by increased inferred hygroscopicity. Organic-rich particles generally showed pronounced pSNR increases with limited or parameter-dependent hygroscopicity changes. BC particles with weaker organic signatures had substantially higher inferred hygroscopicity in aged-like than fresh-like populations during NPs. Their aged-like counterparts during ECPs showed further pSNR and size increases but little additional hygroscopicity enhancement.

Alongside these compositional differences, the coupling among gSNR, pSNR, and nSNR varied with relative humidity (RH). Dry conditions (RH < 55%) were characterized by weaker correlations and more pronounced differences among particle classes. At intermediate RH (55% to 85%), pSNR and D_{va} exhibited nonlinear changes, whereas high RH (> 85%) featured stronger correlations and smaller particle-type contrasts. These patterns are consistent with water uptake and multiphase processing contributing to the RH-dependent of sulfur-nitrogen partitioning, highlighting the environmental context of the observed compositional responses.

These findings suggest that air-quality models may benefit from considering particle-type heterogeneity and RH-dependent processing in gas-particle partitioning and

secondary aerosol formation. Their relevance extends to future emission scenarios with declining gSNR, under which changes in particle-phase sulfur-nitrogen balance may be accompanied by differing hygroscopicity responses among particle populations. Associated changes in particle size, hygroscopicity, and mixing state could influence cloud condensation nuclei (CCN) activation, humidity-dependent light scattering, and aerosol radiative effects (Matsui et al., 2018; Rosenfeld et al., 2014; Titos et al., 2021; Xu et al., 2021). The observed relationships provide a physical basis for investigating these atmospheric consequences across particle types and humidity conditions under evolving emission controls.

Data availability. All data supporting the findings of this study are available within the paper and its Supplementary Information. Additional data related to this paper are available on request by contacting the corresponding author Xinlei Ge (xinlei@seu.edu.cn) and Junfeng Wang (wangjunfeng@nuist.edu.cn).

Supplement. The supplement related to this article is available online at: XXX.

Author contributions. X.G., M.C., and J.W. conceptualized the study and designed the research. Y.D. and S.Z. conducted the field experiments. Y.D. and J.W. analyzed the data and prepared the original draft of the manuscript. X.G., M.C., H.L., Y.W., Y.Z., and M.W. reviewed and edited the manuscript.

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