

**Assessment of Diverse Control Measure Effectiveness During
Successive PM_{2.5} Pollution Episodes in Central China:
Implications for Regional Heavy Pollution Mitigation Strategies**

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Text S1. Uncertainty analysis of daily temporal allocation.

The baseline monthly emissions, emission factors, and coal-substitution factors were treated as fixed inputs, and the uncertainty analysis focused on the daily temporal allocation derived from monitored and proxy activity indicators. Following the reliability-based uncertainty classification of An et al. (An et al., 2021), coefficients of variation (CVs) of 18% were assigned to daily industrial operating rates, road-traffic activity, and non-road construction-machinery activity, whereas a CV of 34% was assigned to the indirectly derived residential-coal proxy. In each realization, source-specific daily activity coefficients were perturbed using mean-one lognormal multiplicative factors and subsequently renormalized within each calendar month to preserve the corresponding monthly source-specific emissions. A total of 10,000 Monte Carlo realizations were performed. For each realization, episode-mean NO_x and primary $\text{PM}_{2.5}$ emissions and their relative changes from E1 to E2 were calculated. Uncertainty was represented by the median, the 2.5th–97.5th percentile uncertainty interval, and the proportion of realizations yielding a positive E2-to-E1 change. The adopted CVs represent literature-informed assumptions based on data-source reliability rather than instrument-specific measurement uncertainties. The resulting uncertainty distributions are presented in Fig. S5, and the corresponding numerical summaries are provided in Table S8. The deterministic estimates were closely aligned with the Monte Carlo medians, indicating that the uncertainty treatment did not materially alter the central estimates.

Text S2. Meteorological Screening Procedure

Meteorological conditions play a critical role in the accumulation, transport, and chemical transformation of PM_{2.5}. To isolate the impacts of emission changes from meteorological variability, meteorologically comparable periods were selected from the two pollution episodes (E1 and E2). Similar meteorological normalization or screening approaches have been widely applied in previous studies to evaluate emission-driven changes in air pollution by controlling for meteorological influences (Zhu et al., 2021). Four key meteorological parameters that strongly influence PM_{2.5} evolution were considered, including temperature (T), relative humidity (RH), wind speed (WS), and precipitation. These parameters represent the effects of atmospheric dispersion, aerosol hygroscopic growth, and wet scavenging processes, respectively. The screening criteria were established based on the observed meteorological characteristics during the two episodes and are summarized (Table S3).

Periods satisfying all meteorological constraints were identified as meteorologically comparable sub-periods and were defined as E1_met and E2_met. Specifically, E1_met corresponded to December 22-28, 2023, while E2_met corresponded to January 8-12, 2024. Compared with the original episode periods, the meteorological screening reduced the differences in atmospheric conditions between E1 and E2, providing a more comparable basis for evaluating emission-related impacts.

Despite the constrained meteorological conditions, substantial differences in PM_{2.5} pollution characteristics remained between E1_met and E2_met. As shown in Table S4 and Fig. S2, the average PM_{2.5} concentration increased from 84.6 $\mu\text{g}/\text{m}^3$ during E1_met to 117.3 $\mu\text{g}/\text{m}^3$ during E2_met, corresponding to an increase of 38.6%. Meanwhile, nitrate and organic matter concentrations increased by 48.7% and 25.0%, respectively, and the nitrate mass fraction increased by 4.0 percentage points. These consistent enhancements under meteorologically comparable conditions indicate that the stronger pollution during E2 was primarily associated with changes in anthropogenic emissions rather than meteorological variability alone.

Text S3. Estimation of Daytime and Nighttime Nitrate Formation

Potentials

Daytime and nighttime nitrate formation potentials were estimated separately because their dominant chemical pathways differ. Previous observational and modeling studies have shown that daytime nitrate formation is generally associated with the gas-phase oxidation of NO_2 by OH, whereas nighttime nitrate formation is strongly influenced by NO_3 - N_2O_5 chemistry, particularly the heterogeneous hydrolysis of N_2O_5 (Zhang et al., 2024; Lin et al., 2025; Ma et al., 2023). Hourly NO_2 and O_3 concentrations were obtained from the provincial air-quality monitoring network, while temperature and surface downward shortwave radiation (SSRD) were obtained from observations and the ERA5 reanalysis dataset. Hours with SSRD greater than zero were classified as daytime, whereas hours with SSRD equal to zero were classified as nighttime. Because direct measurements of OH and N_2O_5 were unavailable, the following calculations were used to characterize the relative chemical conditions conducive to nitrate formation rather than absolute particulate NO_3^- production rates.

During daytime, HNO_3 formation is driven by the gas-phase oxidation of NO_2 by OH, the corresponding HNO_3 production rate is generally expressed as:



$$P_{\text{day}} = k_{\text{OH}+\text{NO}_2} [\text{OH}] [\text{NO}_2] \quad (\text{S1})$$

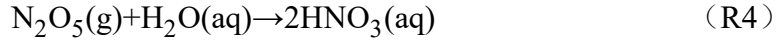
Because direct OH measurements were unavailable, the empirical parameterization developed by Tan et al. was used as the methodological basis for characterizing the temporal variation in OH concentrations (Tan et al., 2025):

$$[\text{OH}] = a * (j\text{O}^1\text{D})^\alpha \frac{(j\text{NO}_2)^\beta * (b\text{NO}_2 + 1)}{c\text{NO}_2^2 + d\text{NO}_2 + 1} \quad (\text{S2})$$

Because $j(\text{O}^1\text{D})$ is primarily controlled by incident solar radiation, hourly surface downward shortwave radiation (SSRD) from ERA5 was used as a proxy for its relative temporal variation. Accordingly, the daytime nitrate production potential was approximated as:

$$P_{\text{day}} \propto \text{SSRD}(t)[\text{NO}_2(t)] \quad (\text{S3})$$

During nighttime, nitrate formation is driven by the oxidation of NO_2 by O_3 , followed by the formation and heterogeneous hydrolysis of N_2O_5 .



Because N_2O_5 was not directly measured, we estimated nighttime nitrate formation under steady-state assumptions, following the study by Jeon et al (Jeon et al., 2024).

$$P_{\text{night}}(t) = 2k_{\text{NO}_2+\text{O}_3}(T)[\text{NO}_2(t)][\text{O}_3(t)] \quad (\text{S5})$$

where the temperature-dependent rate coefficient $k_{\text{NO}_2+\text{O}_3}$ was calculated using the Arrhenius expression reported for atmospheric chemical reactions (Seinfeld and Pandis, 2016).

$$k_{\text{NO}_2+\text{O}_3}(T) = 1.2 \times 10^{-13} \exp\left(-\frac{2450}{T}\right) \quad (\text{S6})$$

To facilitate comparisons between E1 and E2, the hourly potentials were expressed as normalized anomalies relative to the mean value over the entire analysis period.

$$A_{\text{day}}(t) = \frac{P_{\text{day}}(t) - \overline{P_{\text{day}}}}{\overline{P_{\text{day}}}}, \text{SSRD} > 0 \quad (\text{S7})$$

$$A_{\text{night}}(t) = \frac{P_{\text{night}}(t) - \overline{P_{\text{night}}}}{\overline{P_{\text{night}}}}, \text{SSRD} < 0 \quad (\text{S8})$$

where $\overline{P}$ denotes the mean nitrate formation potential over the full study period. Positive values indicate stronger-than-average formation conditions, whereas negative values indicate weaker-than-average conditions. These pathway-specific indices are intended for relative comparisons between the two episodes and should not be interpreted as a comprehensive atmospheric oxidation capacity index or as absolute particulate nitrate production rates.

Tables and Figures

Table S1. Selection of representative stations in Henan Province.

Regional Location	City	Reasons for Selection	Monitoring Sites
North Henan	Anyang	Key industrial city in Northern Henan (steel, coking), where particulate matter composition is significantly influenced by industrial emissions.	Anyang EPA Station (Nation-Controlled)
Central Henan	Zhengzhou	Provincial capital. Prominent mixed pollution from traffic, industry (automobile manufacturing, building materials), and residential sources; represents the complex pollution characteristics of the Central Plains urban agglomeration.	Atmospheric Aerosol Monitoring Station (Province-Controlled)
South Henan	Xinyang	Agricultural eco-region in Southern Henan. Particulate matter is significantly influenced by biomass burning, secondary aerosols, and regional transport (from Hubei direction).	Xinyang Museum Station (Province-Controlled)
West Henan	Sanmenxia	Mountainous city in Western Henan. Influenced by coal transportation, mining, and topographic trapping of pollutants; reflects cross-provincial pollution and topographic effects.	Sanmenxia EPA Station (Province-Controlled)
East Henan	Zhoukou	Agricultural area on the Eastern Henan Plain. Primary particulate matter sources are dust and straw burning, influenced by transport from neighboring provinces (Shandong, Anhui).	Zhoukou Chuanhui Govt. Station (Province-Controlled)

Table S2. The reference oxygen content (cr_{O_2}) of individual industries. An industrial facility is considered in operation if the oxygen content is below cr_{O_2} .

Major facilities of industry	Reference oxygen content
Glass - Kiln	18
Iron and steel - Pellets	19.9
Iron and steel - Sintering machine	19.9
Heating - Boiler	18
Chemical - Boiler + coal burning	18
Thermal power + coal	18
Coking – Coking furnace	19
Refining petrochemical - atmospheric and vacuum	15
Aluminum industry - Roasting kilns	18
Aluminum industry - electrolyzer	21.2
Refractories - Furnaces	20
Lime - Lime kiln	19
Cement – Cement kiln	16
Ceramics – Ceramics kiln	19.5
Colored - roasting kiln	18
Brick – Brick kiln	20.5

Table S3. Meteorological screening criteria

Meteorological Factor	Screening Criteria
Wind Speed	≤ 2.5 m/s (Suppresses horizontal dispersion of pollutants)
Relative Humidity	50% - 80% (Excludes interference from hygroscopic growth and wet deposition)
Temperature	-5°C to 6°C (Covers the thermodynamic characteristics of both periods)
Precipitation	Exclude periods with precipitation > 5 mm (Avoids scavenging effects)

Table S4. Comparison of PM_{2.5} chemical components and related environmental indicators under meteorologically consistent conditions (E1_met vs. E2_met)

Environmental Metric	E1_met	E2_met	Net Percentage Change
Average PM _{2.5} Concentration	84.6µg/m ³	117.3µg/m ³	+38.6%
Nitrate (NO ₃ ⁻) Average Mass Concentration	30.3 µg/m ³	45.1 µg/m ³	+48.7%
Nitrate Mass Fraction in PM _{2.5}	38.2%	42.1%	+4.0 percentage points
Organic Matter (OM) Mass Concentration	16 µg/m ³	20 µg/m ³	+25.0%
Organic Matter Fraction in PM _{2.5}	20.2%	18.7%	-1.5 percentage points
Peak NO ₂ Concentration	67.1 µg/m ³	85.8 µg/m ³	+28.0%
Nitrate Formation Rate Anomaly (Peak)	1.8	2.8	+52.0%

Table S5. Configuration of the WRF-CAMx modeling system.

Model	Configuration item	Setting
WRF	Dynamic core	Advanced Research WRF (ARW)
WRF	Simulation mode	31 h segments initialized at 12:00 UTC
WRF	External meteorological input interval	6 h
WRF	Number of domains	2
WRF	Nesting method	Two-way nesting
WRF	Horizontal grid resolution	36 km for d01; 12 km for d02
WRF	Grid dimensions	d01: 210 × 200; d02: 133 × 145
WRF	Parent-grid ratio	1:3
WRF	Map projection	Lambert conformal
WRF	Reference latitude and longitude	35.0°N, 110.0°E
WRF	True latitudes	24.0°N and 46.0°N
WRF	Standard longitude	110.0°E
WRF	Vertical layers	35 eta levels
WRF	Model top	5000 Pa (50 hPa)
WRF	Model time step	120 s
WRF	Output interval	60 min
WRF	Microphysics	WRF single-moment three-class scheme (WSM3)
WRF	Longwave radiation	Rapid Radiative Transfer Model (RRTM)
WRF	Shortwave radiation	Dudhia scheme
WRF	Radiation call interval	30 min
WRF	Surface-layer scheme	Revised MM5 similarity scheme
WRF	Land-surface model	Noah land-surface model

Model	Configuration item	Setting
WRF	Planetary boundary-layer scheme	Yonsei University scheme
WRF	Cumulus parameterization	Kain–Fritsch scheme for both domains
WRF	Cumulus call interval	5 min
WRF	Number of soil layers	4
WRF	Land-use categories	21
WRF	Urban canopy option	Enabled for d01; disabled for d02
WRF	Analysis nudging	Not applied
CAMx	Model version	CAMx version 7.10
CAMx	Simulation period	December 12, 2023-January 24, 2024
CAMx	Model time zone	UTC
CAMx	Run mode	Consecutive daily simulations with restart files
CAMx	Number of grids	2
CAMx	Master-grid resolution	36 km
CAMx	Master-grid dimensions	200 × 160
CAMx	Nested-grid resolution	12 km
CAMx	Nesting ratio	1:3
CAMx	Nested-grid location in master grid	I = 102-140; J = 77-120
CAMx	Map projection	Lambert conformal
CAMx	Reference latitude and longitude	35.0°N, 110.0°E
CAMx	True latitudes	24.0°N and 46.0°N
CAMx	Number of vertical layers	20
CAMx	Maximum time step	15 min
CAMx	Meteorological input interval	60 min

Model	Configuration item	Setting
CAMx	Emission input interval	60 min
CAMx	Output interval	60 min
CAMx	Chemistry-parameter file	SAPRC07_CF2
CAMx	Chemistry solver	Euler backward iterative solver
CAMx	Advection solver	Piecewise parabolic method
CAMx	Photolysis rates	TUV version 4.8
CAMx	Ozone-column input	Monthly ozone-column field
CAMx	Dry deposition	Zhang03 scheme
CAMx	Wet deposition	Enabled
CAMx	Super-stepping	Enabled
CAMx	Plume-in-grid submodel	Not applied
CAMx	Gridded emissions	Enabled
CAMx	Point-source input	Disabled; emissions were provided through gridded inputs
CAMx	Initial conditions	Species-specific constant concentrations generated using ICBCPREP
CAMx	Lateral boundary conditions	Species-specific constant concentrations generated using ICBCPREP
CAMx	IC/BC input format	TOPCON-format concentration file (tc_1.use)
CAMx	IC/BC grid configuration	200 × 160 × 20 layers at 36 km resolution
CAMx	Output format	NetCDF
CAMx	Output species	All modeled species
CAMx	Dynamic anthropogenic emissions	Daily resolved gridded emissions developed in Section 2.2

Table S6. General information of episodes E1 and E2

Air pollution events	Duration (days)	Number of heavy polluted cities	Number of severe polluted cities	Average PM _{2.5} conc. (μg/m ³)	Maximum PM _{2.5} daily conc. (μg/m ³)	Maximum PM _{2.5} hourly conc. (μg/m ³)
E1	13	16	0	104	230 (Zhoukou, Dec. 29 th)	263 (Sanmenxia, 12:00 on Dec. 29 th)
E2	7	17	12	142	312 (Kaifeng, Jan. 13 th)	398 (Kaifeng, 22:00 on Jan. 13 th)

Table S7. Emergency response to pollution sources during E1 and E2 (comparison of industrial source operating rates and traffic flow changes)

Pollution source / Activity		% Change (E2 vs. E1)
Industrial source operating rate	Thermal power and heating	+3%
	Non-interruptible industries	-3%
	Interruptible industries	+23%
	Iron and steel -sintering	-32%
	Cement	+44%
	Bricks and tiles	+25%
	Refractory materials	+40%
	Lime kilns	+65%
Heavy-duty truck traffic flow		+20%
Heavy cargo flow at provincial border checkpoints		+16%
Operating hours of non-road machinery		+21%

Table S8. Monte Carlo uncertainty in episode-mean emissions and their changes from E1 to E2.

Pollutant	Deterministic E1 (t d ⁻¹)	Deterministic E2 (t d ⁻¹)	Deterministic change (%)	MC median change (%)	95% UI (%)	(P($\Delta > 0$)) (%)
NO _x	2188.5	2476.5	13.2	13.2	2.9– 24.7	99.5
Primary PM _{2.5}	653.5	742.1	13.6	13.3	–4.5– 33.9	92.7

Note: UI denotes the 2.5th–97.5th percentile uncertainty interval, and Δ denotes the percentage change from E1 to E2. P($\Delta > 0$) represents the percentage of Monte Carlo realizations yielding a positive change. Baseline monthly emissions, emission factors, and coal-substitution factors were held fixed.

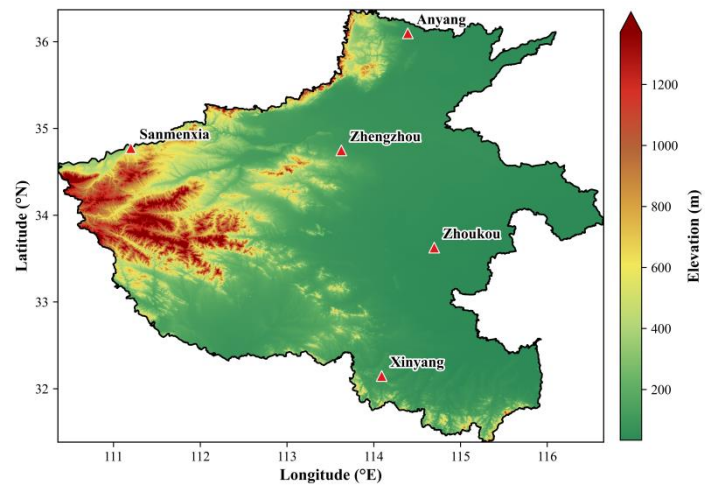


Figure S1. Digital elevation model (DEM) of Henan province and distribution of representative urban sites.

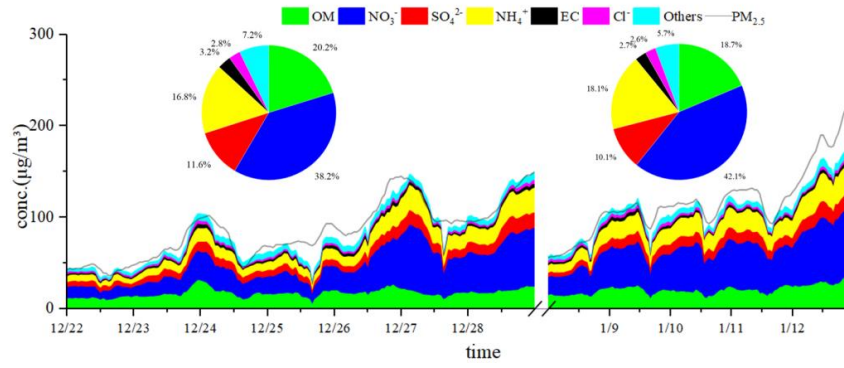


Figure S2. Trends in PM_{2.5} chemical components and their mass proportions in Henan Province during E1_met and E2_met sub-periods after meteorological filtering.

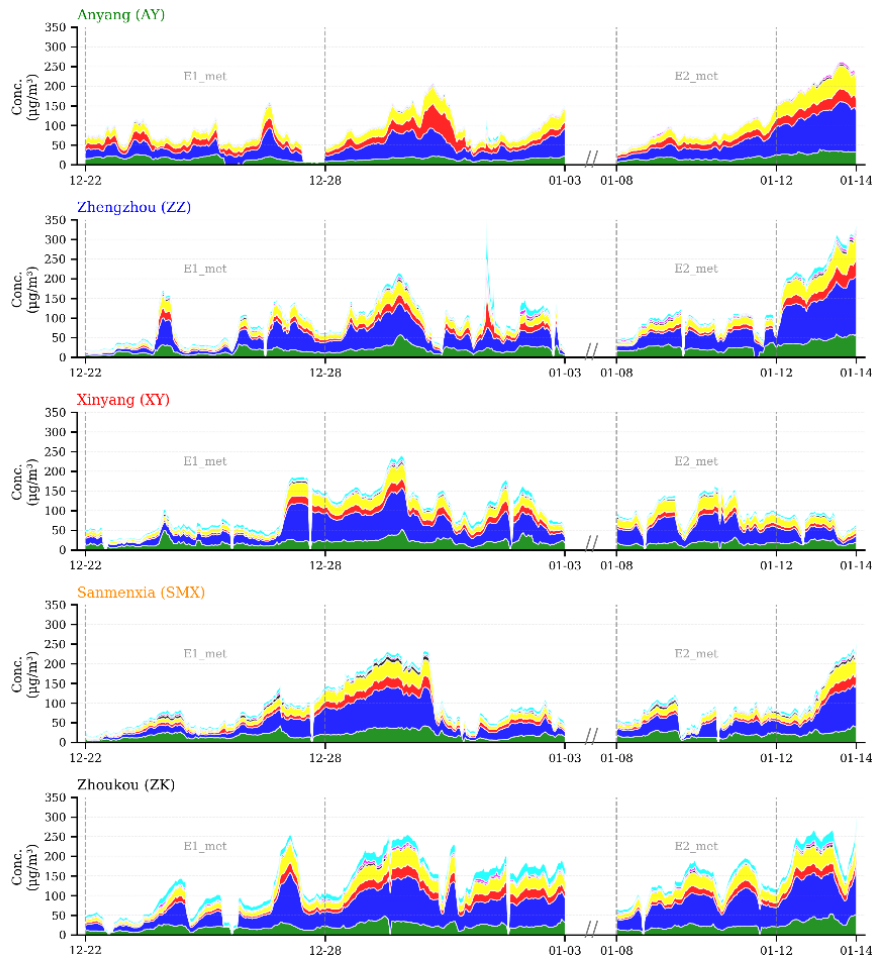


Figure S3. Temporal variations of PM_{2.5} components at representative monitoring sites in Henan Province during the E1 and E2 episodes.

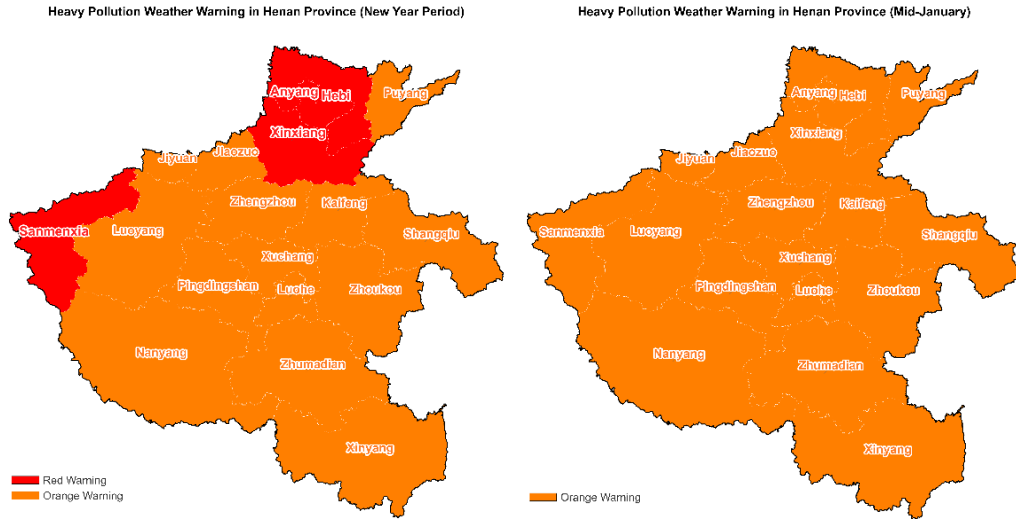


Figure S4. Heavy pollution warning maps for E1 and E2.

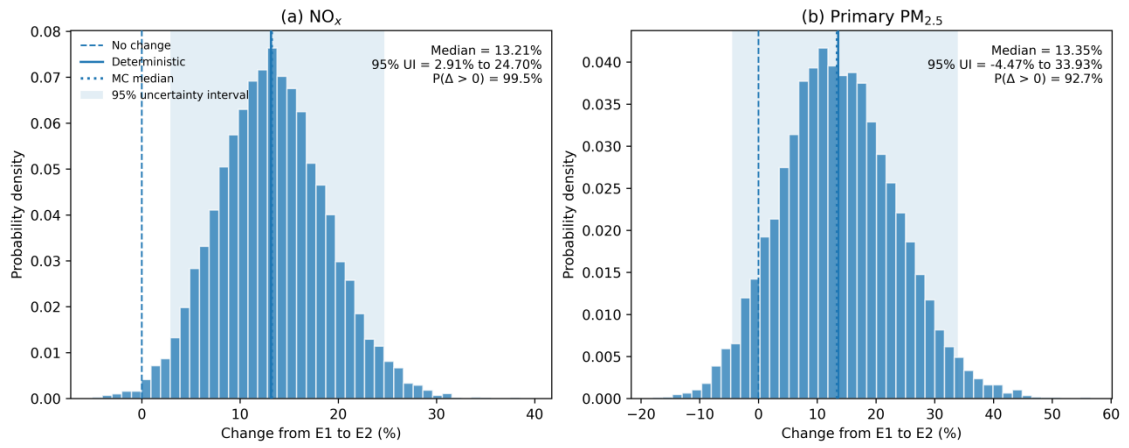


Figure S5. Monte Carlo distributions of the E2-to-E1 changes in (a) NO_x and (b) primary $\text{PM}_{2.5}$ emissions. Solid and dotted lines denote the deterministic estimates and Monte Carlo medians, respectively; the dashed line indicates zero change, and shading represents the 95% uncertainty interval.

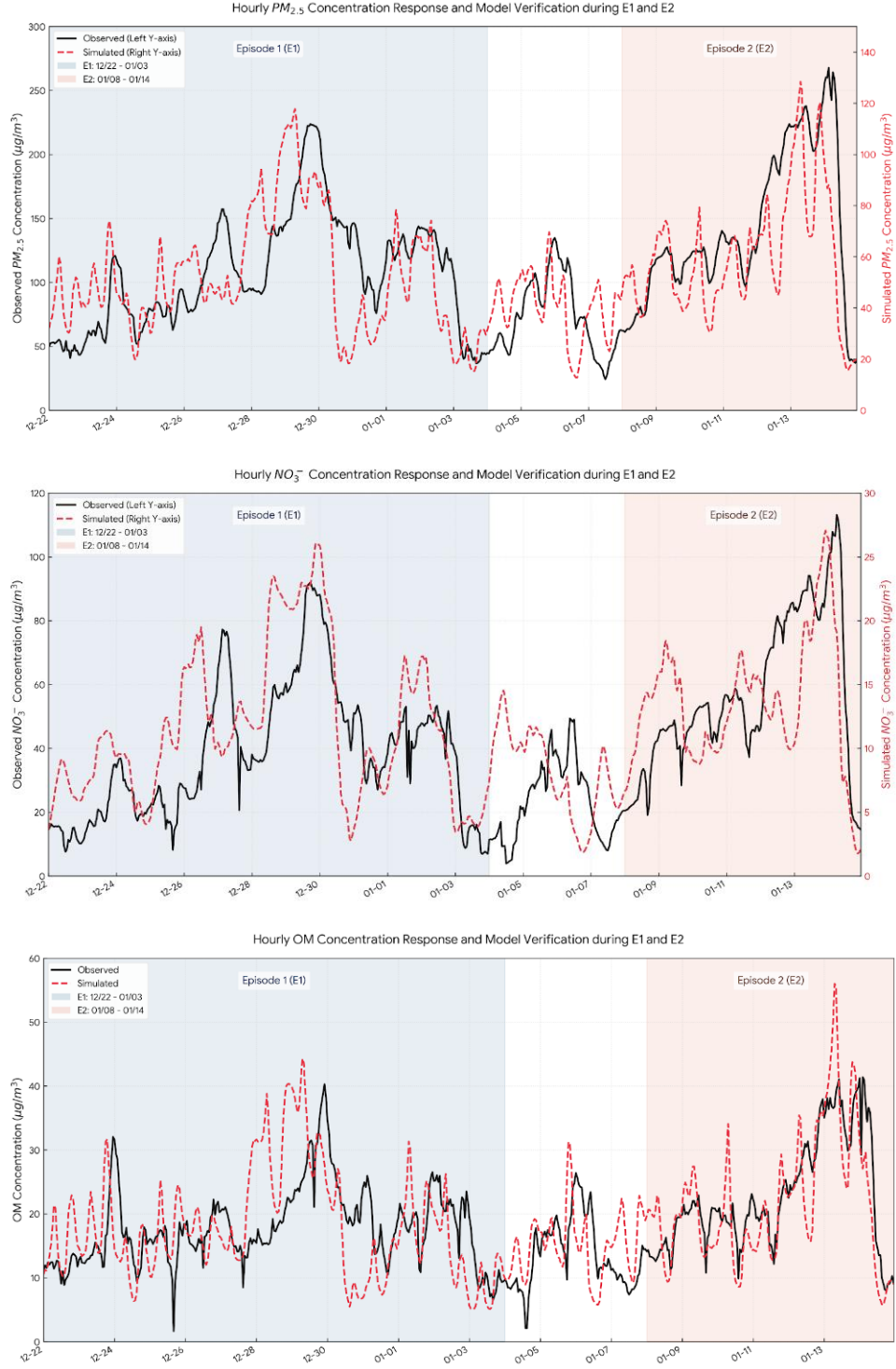


Figure S6. Observed versus simulated concentrations of $\text{PM}_{2.5}$, nitrate (NO_3^-), and organic matter (OM) during two consecutive heavy pollution events in Henan Province.

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