

Anthropogenic emissions drive elevated HCl and particulate chloride in polluted urban air

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Supplementary Text 1. Model Configuration

In this work, the physical and chemical processes governing chloride formation were investigated through simulations using the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) (Version 4.2.2)¹. The model configuration comprised the Grell–Freitas (GF)² method for cumulus parameterization, the Rapid Radiative Transfer Model for GCMs (RRTMG)³ for both shortwave and longwave radiation, and the Lin et al.⁴ microphysics scheme for cloud and precipitation processes. The Lin microphysics scheme was selected based on prior regional evaluations indicating improved precipitation and cloud representation over India compared to more complex schemes.⁵ The Yonsei University planetary boundary layer (PBL)⁶ scheme was used to treat boundary layer dynamics, while the Noah land surface model⁷ was used to depict surface–atmosphere interactions. MOZART (Model for Ozone and Related Tracers)⁸ was used to simulate gas-phase chemistry, and MOSAIC (Model for Simulating Aerosol Interactions and Chemistry)⁹ was used to represent aerosol processes.

The European Centre for Medium-Range Weather Forecasts (ECMWF) provided the ERA5 reanalysis¹⁰, which had a 6-hourly temporal resolution and a spatial resolution of $0.25^\circ \times 0.25^\circ$, from which meteorological initial and boundary conditions were obtained. The Emissions Database for Global Atmospheric Research (EDGAR v6.1)^{11,12}, which offers monthly mean data at $0.1^\circ \times 0.1^\circ$ resolution, was used to obtain anthropogenic emissions with reference to 2018. The Model of Emissions of Gases and Aerosols from Nature (MEGAN, version 2.1)¹³ was used to generate biogenic emissions, and the NCAR Fire Inventory (FINN, version 2.5)¹⁴ was used to incorporate fire emissions. The Whole Atmosphere Community Climate Model (WACCM)¹⁵ provided the initial and chemical lateral boundary conditions.

The WACCM model employs the Modal Aerosol Model (MAM4)¹⁶, which represents aerosols using lognormal modes (Aitken, accumulation, coarse, and primary carbon), whereas MOSAIC uses a sectional representation with discrete size bins. In this study, the 4-bin MOSAIC configuration was used. The mapping of aerosol mass and number concentrations from MAM4 to MOSAIC bins was performed using community tools provided by UCAR (<https://www2.aom.ucar.edu/wrf-chem/wrf-chem-tools-community>). This mapping redistributes species based on the overlap between the modal size

distributions in MAM4 and the size ranges of MOSAIC bins, ensuring conservation of aerosol mass across species and modes. As a result, each MOSAIC bin receives contributions from multiple MAM4 modes through weighted linear combinations. The detailed mapping coefficients used in this study are provided in Supplementary Table 2.

Supplementary Text 2. Evaluation of NH₃ Prediction

The formation of particulate chloride (Cl⁻) through ammonium chloride (NH₄Cl) depends not only on the availability of hydrogen chloride (HCl), but also on the abundance of ammonia (NH₃), which neutralizes acidic species and drives gas-particle partitioning. Therefore, biases in NH₃ could potentially influence simulated particulate chloride concentrations.

Model evaluation against observations indicates that NH₃ is generally well reproduced or overestimated across all three seasons (Supplementary Fig. 16)¹⁷. During the post-monsoon period, NH₃ is overpredicted (mean: ~37.24 µg m⁻³ modeled vs ~28.83 µg m⁻³ observed). In winter, modeled and observed concentrations are in close agreement (~34.39 vs ~34.12 µg m⁻³). During summer, NH₃ is again slightly overestimated (~25.60 vs ~23.30 µg m⁻³).

The consistent agreement or overprediction of NH₃ suggests that the system is predominantly ammonia-rich rather than ammonia-limited, particularly in post-monsoon and summer conditions. Therefore, the underestimation of particulate chloride in the model, especially during summer, is unlikely to be driven by insufficient NH₃, and instead points toward uncertainties in the thermodynamic treatment of HCl partitioning, including the effective equilibrium constant used in the model.

Supplementary Text 3. Impact of HCl emissions on simulated chemical fields

The comparison of model-simulated chemical fields with and without anthropogenic HCl and particulate chloride (pCl) emissions provides a quantitative assessment of the baseline model performance and the impact of these emissions on aerosol composition. Supplementary Fig. 17 presents the evaluation of key species (NH₃, PM_{2.5}, NH₄⁺, NO₃⁻) across three seasons (post-monsoon, winter, and summer), where the default simulation excludes HCl and pCl emissions, and the revised simulation includes them.

NH₃ concentrations are consistently high across all seasons (Supplementary Fig. 17a-c), indicating an NH₃-rich atmospheric environment. The model overestimates NH₃ compared to observations, consistent with previous findings (Pawar et al., 2023)¹⁷. The overestimation is higher in the default simulation and is reduced in the revised simulation, with mean decreases of 1.14 µg m⁻³ (post-monsoon), 1.51 µg m⁻³ (winter), and 4.29 µg m⁻³ (summer) at the IIT Delhi grid. This reduction is attributed to the enhanced consumption of NH₃ through reactions with HCl, leading to increased formation of ammonium chloride.

PM_{2.5} concentrations in the default simulation agree well with observations (Supplementary Fig. 17d-f) during the post-monsoon and winter seasons but are overestimated during summer. The revised simulation shows a slight increase in PM_{2.5} across seasons, reflecting the additional contribution of particulate chloride from the inclusion of HCl and pCl emissions.

NH₄⁺ concentrations increase in the revised simulation across all seasons, with mean enhancements of 5.64 µg m⁻³ (post-monsoon), 4.9 µg m⁻³ (winter), and 1.45 µg m⁻³

(summer), consistent with increased ammonium chloride formation (Supplementary Fig. 17g-i). Observed NH_4^+ concentrations during winter remain higher than model predictions, a discrepancy also reported in previous studies¹⁷. It is noted that ACSM measurements were unavailable during the post-monsoon period, and particulate chloride data for this season were obtained from XACT. NO_3^- concentrations show minimal differences between the default and revised simulations across all seasons (Supplementary Fig. 17j-l), indicating that the inclusion of HCl emissions has a limited impact on nitrate chemistry under the conditions considered.

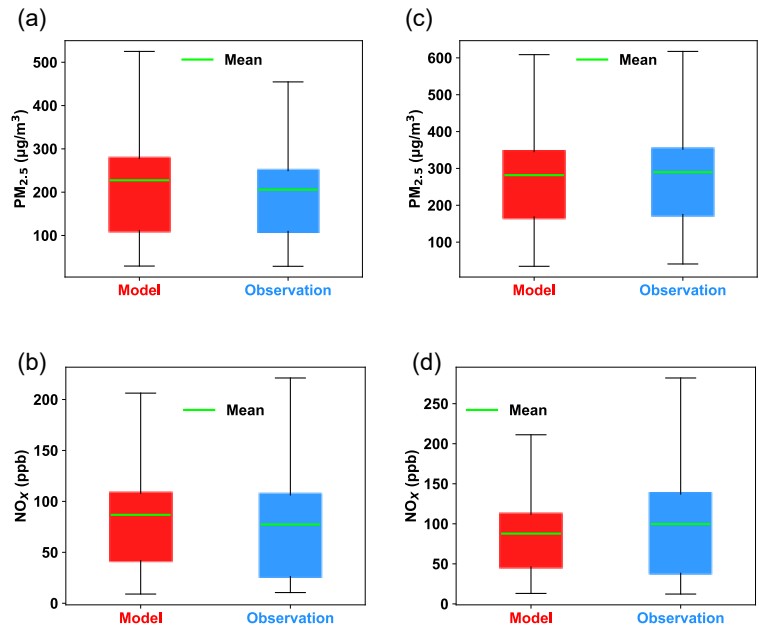
Supplementary Text 4. Diurnal Variation of HCl & Cl^- and number concentrations in Delhi

The diurnal variations of HCl and particulate chloride (Cl^-) are presented in Supplementary Fig. 4. An inverse relationship between HCl and Cl^- is observed across seasons, reflecting gas-particle partitioning between HCl (gas phase) and particulate chloride. During the post-monsoon and winter seasons, HCl concentrations peak during the daytime, whereas Cl^- concentrations are higher during nighttime, with maximum values in the early morning. This behavior is driven by lower temperatures during nighttime and early morning, which favor the partitioning of HCl into the particle phase.

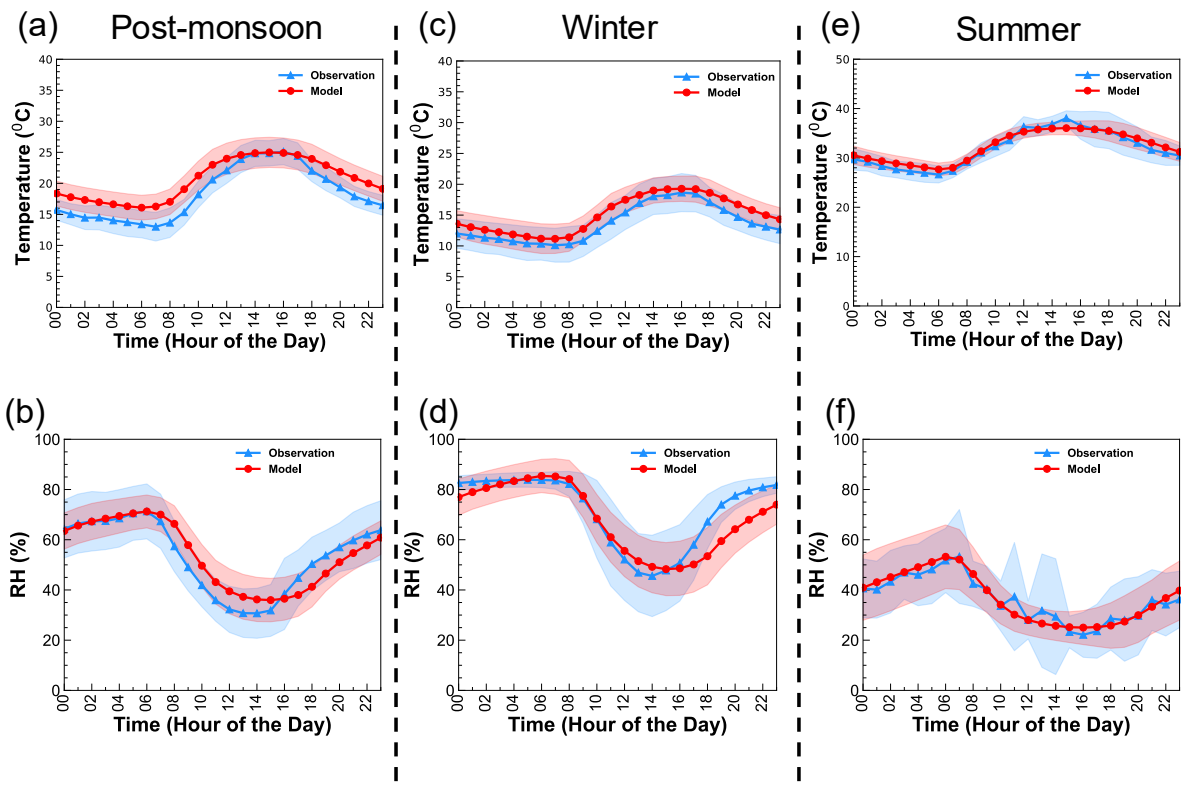
In contrast, during summer, daytime Cl^- concentrations are nearly negligible due to high temperatures and enhanced boundary layer mixing. The diurnal variation of HCl also differs from other seasons, although the total chloride ($\text{HCl} + \text{Cl}^-$) remains relatively consistent (Supplementary Fig. 10). The distinct summer behavior can be attributed to the significantly higher daytime boundary layer height (~2400 m), compared to winter (~700 m) and post-monsoon (~1000 m), leading to dilution of HCl concentrations.

The particle number concentrations in Delhi remain in the range of ~25,000–35,000 cm^{-3} across all seasons (Supplementary Fig. 6), with the highest values during winter and lowest during summer. Aitken mode particles dominate the number concentration throughout the year, while an increased contribution from accumulation mode particles is observed during winter.

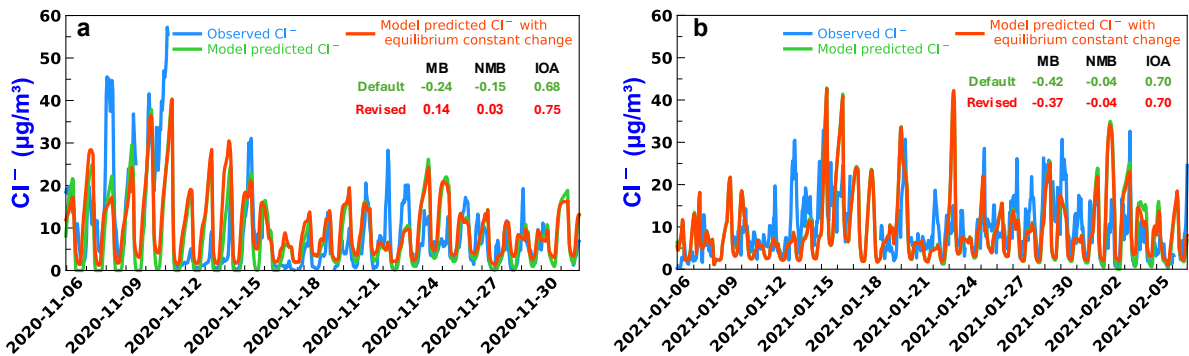
Supplementary Figures



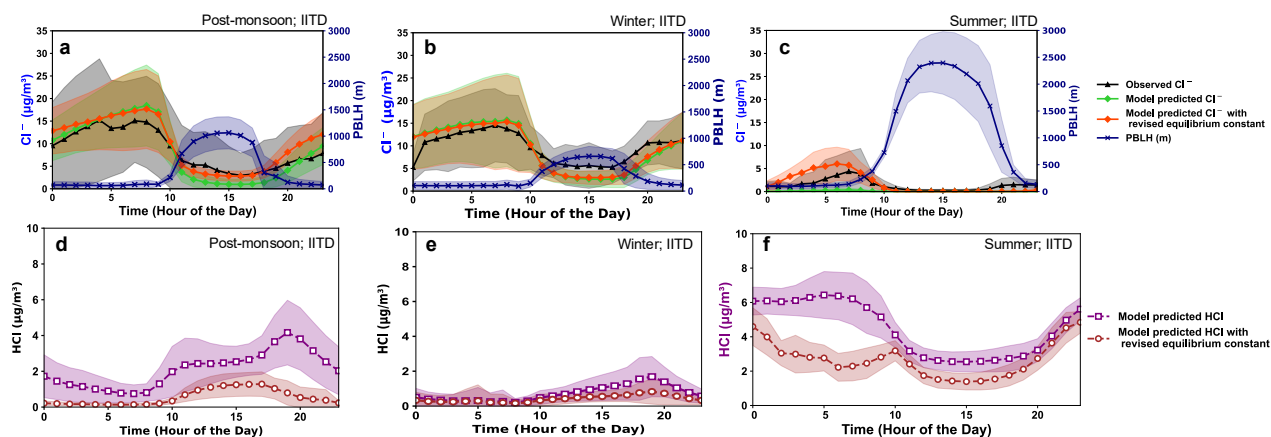
Supplementary Fig. 1 | Box-and-whisker plots of PM_{2.5} and NO_x concentrations for the post-monsoon (a, b) and winter (c, d) seasons, comparing observations and model simulations. Boxes represent the interquartile range with median values, and whiskers indicate data variability.



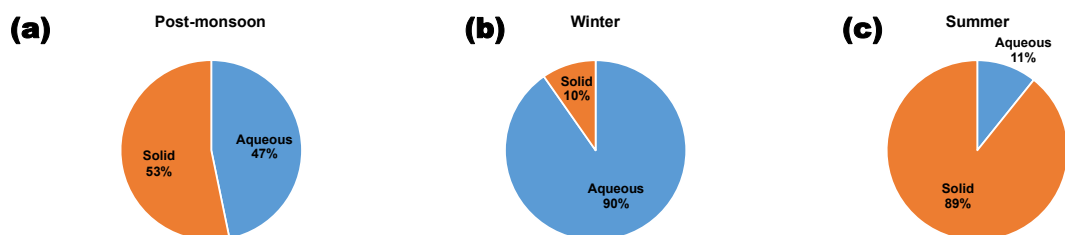
Supplementary Fig. 2 | Meteorological variable diurnal variation. a, b, Temperature and Relative Humidity diurnal in post-monsoon; c, d, Temperature and Relative Humidity diurnal in winter; e, f, Temperature and Relative Humidity diurnal in summer.



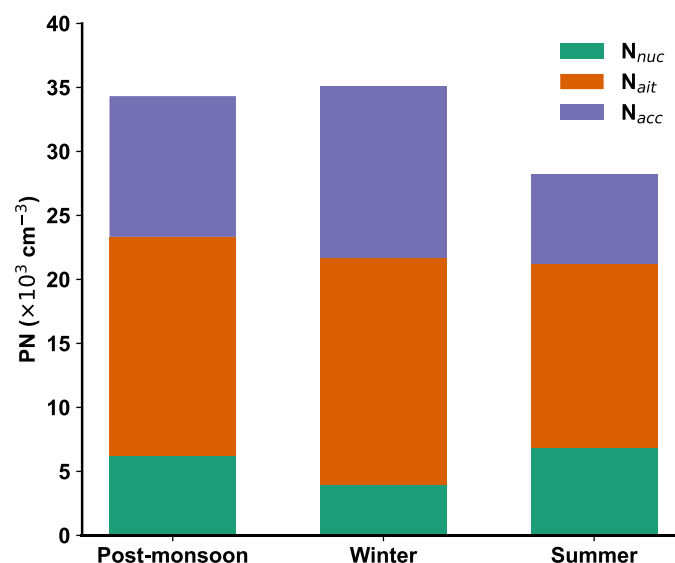
Supplementary Fig. 3 | Time series of particulate chloride concentrations for the post-monsoon (a) and winter (b) seasons, comparing observations and model simulations. Statistical metrics including mean bias (MB), normalized mean bias (NMB), and index of agreement (IOA) are shown in the figure.



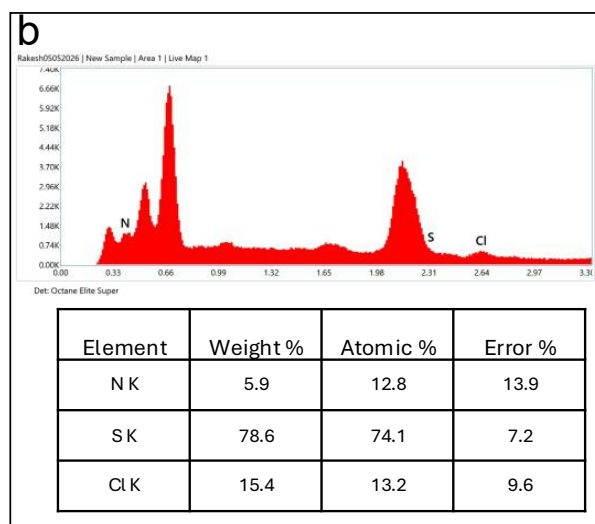
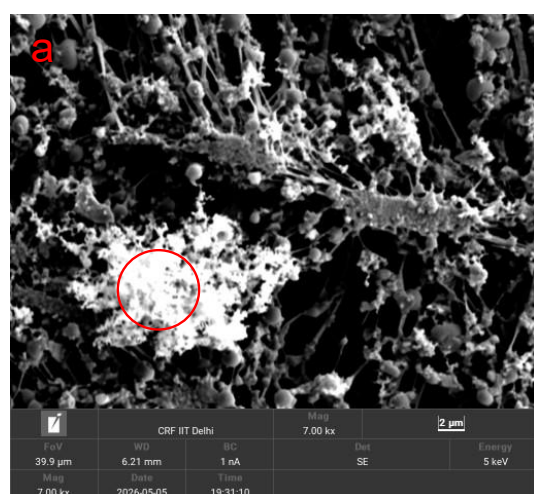
Supplementary Fig. 4 | Diurnal variation of Cl^- and HCl (black – observed particulate chloride; green – base model predicted particulate chloride; red – model predicted particulate chloride with revised equilibrium constant; purple – base model predicted HCl ; brown – model predicted HCl with revised equilibrium constant). a, Cl^- and HCl variation in post-monsoon; b, Cl^- and HCl variation in winter; c, Cl^- , HCl and Planetary Boundary Layer Height (PBLH) (meters) variation in summer (Diurnal of HCl in summer is different compared to other two seasons, which we attribute to the higher PBLH). The diurnal cycle is calculated over the entire date range of the respective season at IIT Delhi site.



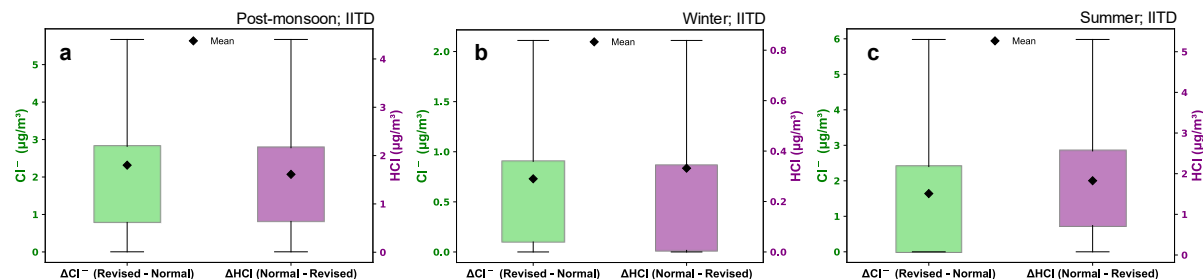
Supplementary Fig. 5 | Model-simulated contributions of particulate chloride in the solid and aqueous phases across seasons: post-monsoon (a), winter (b), and summer (c). Results are derived from sensitivity simulations in which the gas-to-solid equilibrium partitioning constant (K_{gs}) is increased by a factor of 5 to enhance solid-phase formation. This allows assessment of how chloride is redistributed between solid and aqueous phases under modified partitioning conditions.



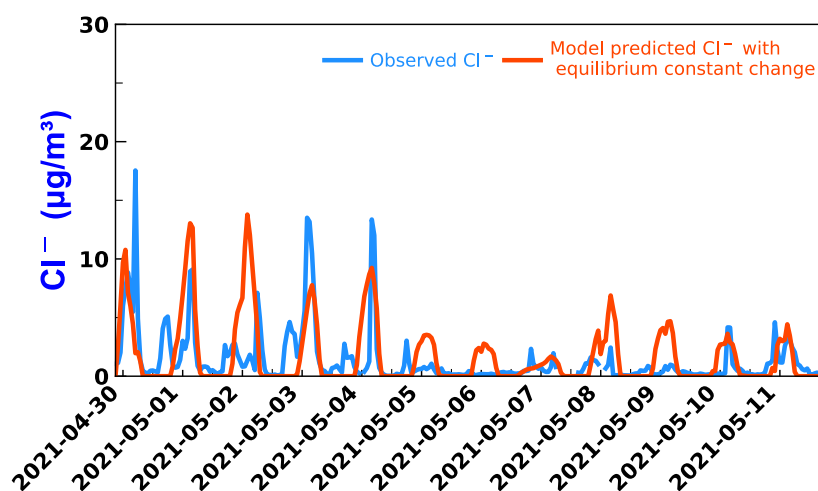
Supplementary Fig. 6 | Seasonal average particle number concentrations in Delhi measured using a scanning mobility particle sizer (SMPS) at IIT Delhi. Green, orange, and purple segments represent nucleation-, Aitken-, and accumulation-mode particles, respectively. The data corresponds to measurements from 2022 and are shown to illustrate typical seasonal particle number concentrations in Delhi rather than the exact model simulation period.



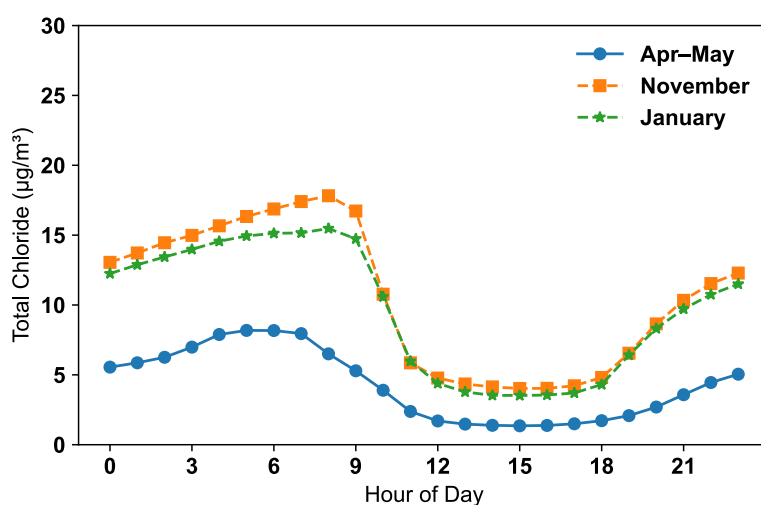
Supplementary Fig. 7 | SEM-EDX analysis of sulfur-rich aerosol particles collected in Delhi during summer. (a) SEM image showing aggregated aerosol particle morphology. The circled region represents the approximate area analyzed using EDX. (b) Corresponding EDX spectrum and elemental composition. Existence of high chlorine in sulfur rich particles indicates the possibility of heterogenous reaction on to the surface of the sulfur particles.



Supplementary Fig. 8 | Seasonal differences between default and revised model simulations of particulate chloride (Cl^-) and HCl at the IIT Delhi site. (a) Post-monsoon; (b) Winter; (c) Summer. For Cl^- , the difference is calculated as revised minus Normal (default) model. For HCl, the difference is calculated as default minus revised model, reflecting the decrease in HCl concentrations in the revised simulation.

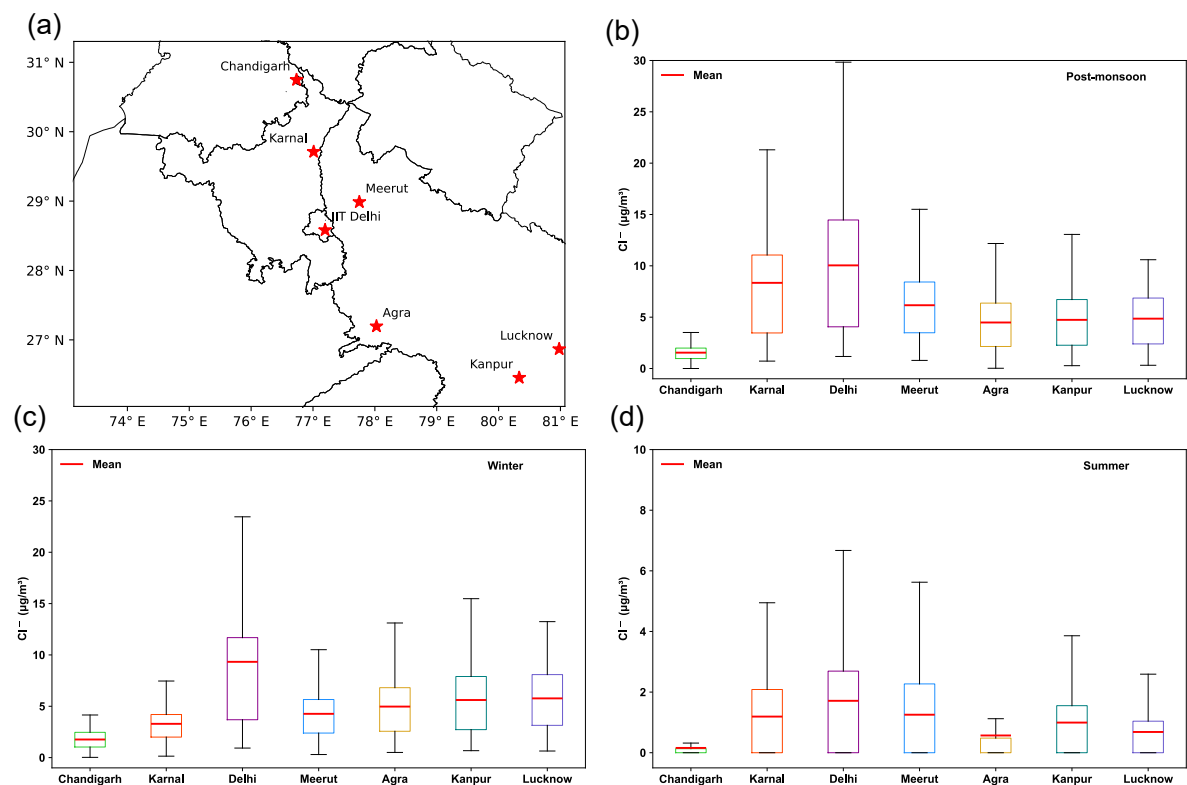


Supplementary Fig. 9 | Time series of particulate chloride for the summer season. The default model predictions are not shown due to negligible concentrations.



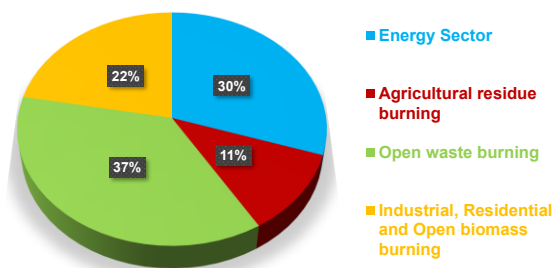
Supplementary Fig. 10 | Diurnal variation of model-predicted total chloride across seasons. Total chloride is calculated as the sum of particulate chloride and HCl (converted to chloride mass using a factor of 35.45/36.46).

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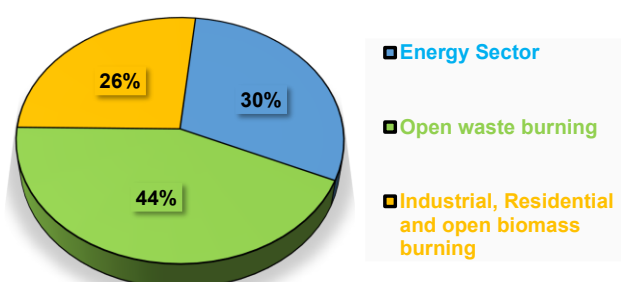
Supplementary Fig. 11 | Particulate chloride concentration over seven major cities in the domain. a, Location of the different cities in the domain; **b,** Box and whiskers plot in post-monsoon; **c,** Box and whiskers plot in winter; **d,** Box and whiskers plot in summer.

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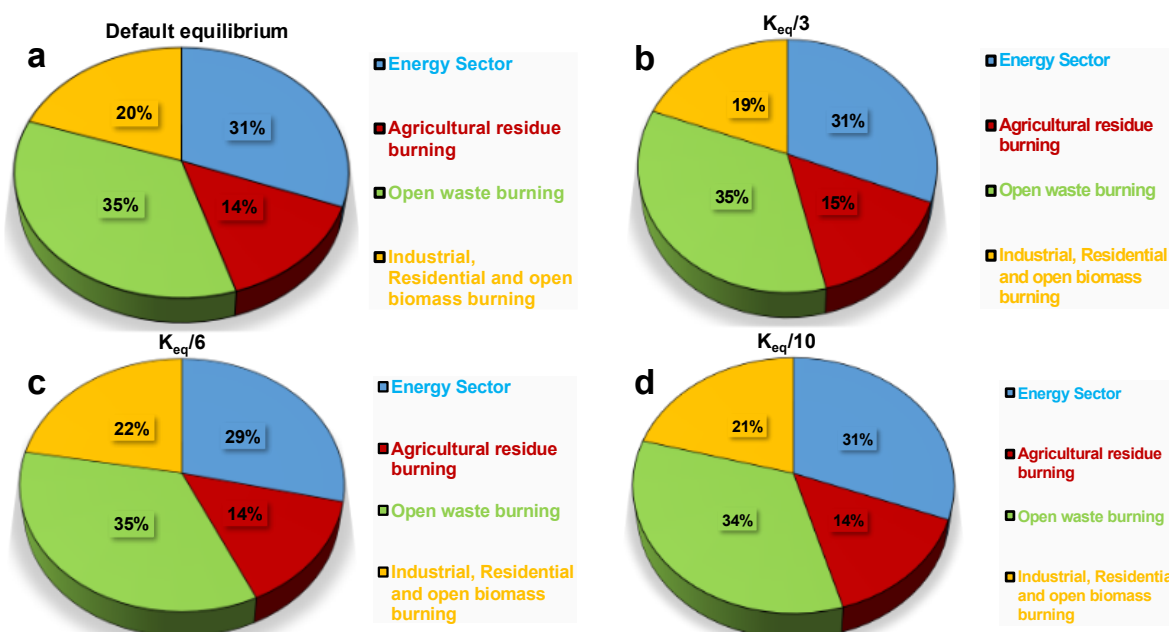


Supplementary Fig. 12 | Source contribution during summer in Delhi

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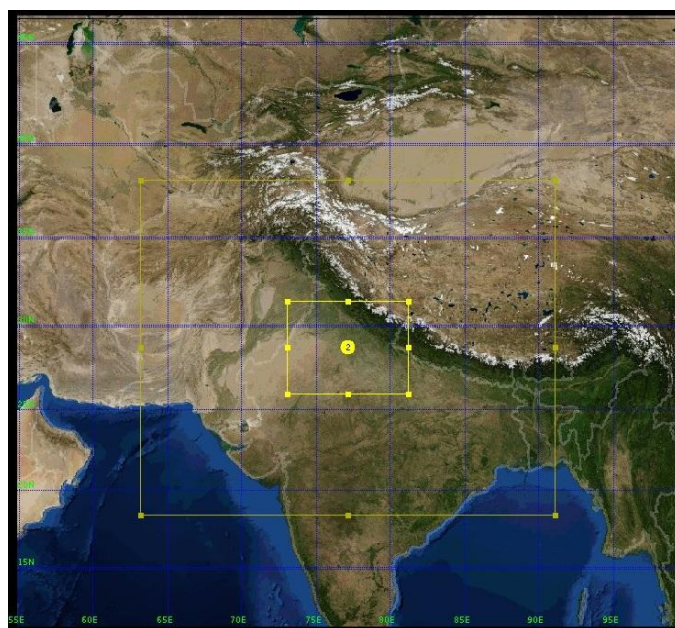


Supplementary Fig. 13 | Source contribution during winter with default model equilibrium in IIT Delhi grid



Supplementary Fig. 14 | Sensitivity of the source apportionment of the different model simulations at IIT Delhi grid. a, Default equilibrium in the model; b, With equilibrium decreased by a factor of three ($K_{eq}/3$); c, With equilibrium decreased by a factor of three ($K_{eq}/6$); d, With equilibrium decreased by a factor of three ($K_{eq}/10$).

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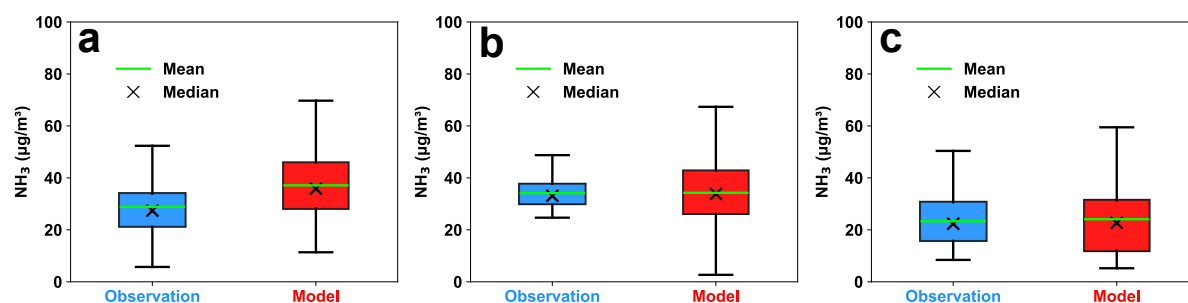


Supplementary Fig. 15 | Model domains used in this study showing two nested domains. The outer domain (d01) and inner domain (d02) cover the broader region and the study area at higher resolution, respectively. Horizontal resolutions are 27 km (d01) and 9 km (d02), with grid dimensions of 100×81 and 88×67 (west-east $\times$ south-north), respectively. The same model configuration was used as in our previous study by Maity et al. (2026)¹⁸.

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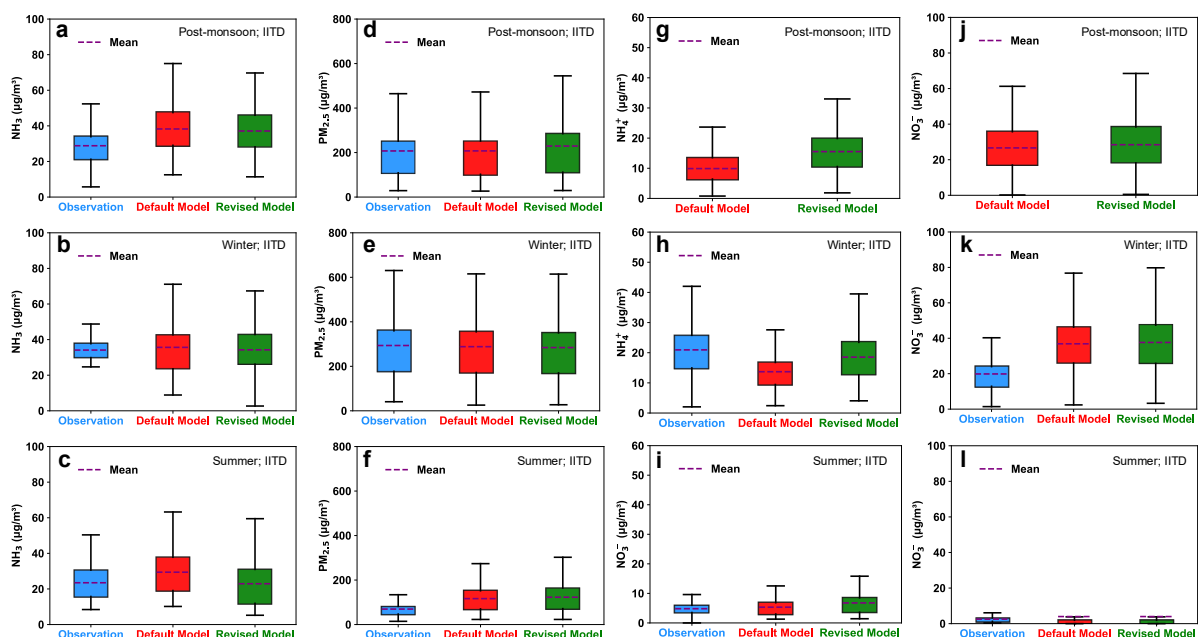
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Supplementary Fig. 16 | Comparison of model predicted NH_3 for the three seasons with observations from CPCB sites averaged over the IIT Delhi grid. a, NH_3 of Post-monsoon. b, NH_3 of Winter. c, NH_3 summer.

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Supplementary Fig. 17 | Comparison of NH_3 , $\text{PM}_{2.5}$, NH_4^+ , and NO_3^- concentrations between observations, default simulation, and revised simulation across post-monsoon, winter, and summer seasons. Box-and-whisker plots illustrate the distribution of each species, highlighting the impact of incorporating anthropogenic HCl and pCl emissions in the revised model.

Supplementary Table 1: Summary of numerical experiments

SL. NO.	Simulation Name	Configuration	Key Modification	Objective
1	Default	Standard WRF-Chem	N.A.	Evaluate baseline model performance
2	Default + anthro. HCl & pCl	anthropogenic HCl and pCl emissions are included	Inclusion of anthropogenic HCl and particulate chloride (pCl) emissions	Assess the impact of anthropogenic HCl and pCl
3	Sensitivity study of Equilibrium constant	Keq/3 + Default + anthropogenic HCl and pCl emissions	Gas to solid equilibrium partitioning changed by a factor of 3	To minimize the bias between the observation and model prediction
4		Keq/6 + Default + anthropogenic HCl and pCl emissions	Gas to solid equilibrium partitioning changed by a factor of 6	To minimize the bias between the observation and model prediction
5		Keq/10 + Default + anthropogenic HCl and pCl emissions	Gas to solid equilibrium partitioning changed by a factor of 10	To minimize the bias between the observation and model prediction

6	Source apportionment	Open waste burning + Default + anthropogenic HCl and pCl emissions + Keq/10	Only open waste burning source is considered	Quantify the impact on north Indian cities
7		Energy + Default + anthropogenic HCl and pCl emissions + Keq/10	Only energy sector is considered	
8		Agricultural residue burning + Default + anthropogenic HCl and pCl emissions + Keq/10	Only agricultural residue burning source is considered	
9		Other sources + Default + anthro. HCl & pCl + Keq/10	Other sources are considered	
10	Sensitivity of source apportionment with equilibrium constant	Open waste burning/Energy/Agricultural residue burning/Others + Default + anthropogenic HCl and pCl emissions + Keq	To assess the impact of gas to particle partition in individual sources	Sensitivity of the sources with gas to particle partitioning
11		Open waste burning/Energy/Agricultural residue burning/Others + Default + anthropogenic HCl and pCl emissions + Keq/3		
12		Open waste burning/Energy/Agricultural residue burning/Others + Default + anthropogenic HCl and pCl emissions + Keq/6		
13		Open waste burning/Energy/Agricultural residue burning/Others + Default + anthropogenic HCl and pCl emissions + Keq/10		
14	Gas-to-solid fraction	Default + anthropogenic HCl and pCl emissions + increasing Keq to a very high value	Increasing Keq to a very high value (~5 orders of magnitude high compared to standard model)	Estimate percentage of gas-to-solid partitioning

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191 **Supplementary Table 2: Mapping of the WACCM species in MOSAIC**

MOSAIC	WACCM
oc_a01	0.1216*pom_a1+0.9886*soa1_a2+0.1216*soa1_a1+0.9886*soa2_a2+0.1216*soa2_a1+0.9886*soa3_a2+0.1216*soa3_a1+0.9886*soa4_a2+0.1216*soa4_a1+0.9886*soa5_a2+0.1216*soa5_a1;1.e9

oc_a02	$0.7618 \cdot \text{pom_a1} + 0.0114 \cdot \text{soa1_a2} + 0.7618 \cdot \text{soa1_a1} + 0.0134 \cdot \text{soa2_a2} + 0.7618 \cdot \text{soa2_a1} + 0.0114 \cdot \text{soa3_a2} + 0.7621 \cdot \text{soa3_a1} + 0.0114 \cdot \text{soa4_a2} + 0.7621 \cdot \text{soa4_a1} + 0.0114 \cdot \text{soa5_a2} + 0.7621 \cdot \text{soa5_a1}; 1.e9$
oc_a03	$0.1164 \cdot \text{pom_a1} + 0.0000 \cdot \text{soa1_a2} + 0.1164 \cdot \text{soa1_a1} + 0.0000 \cdot \text{soa2_a2} + 0.1164 \cdot \text{soa2_a1} + 0.0000 \cdot \text{soa3_a2} + 0.1164 \cdot \text{soa3_a1} + 0.0000 \cdot \text{soa4_a2} + 0.1164 \cdot \text{soa4_a1} + 0.0000 \cdot \text{soa5_a2} + 0.1164 \cdot \text{soa5_a1}; 1.e9$
oc_a04	$0.0002 \cdot \text{pom_a1} + 0.0000 \cdot \text{soa1_a2} + 0.0002 \cdot \text{soa1_a1} + 0.0000 \cdot \text{soa2_a2} + 0.0002 \cdot \text{soa2_a1} + 0.0000 \cdot \text{soa3_a2} + 0.0002 \cdot \text{soa3_a1} + 0.0000 \cdot \text{soa4_a2} + 0.0002 \cdot \text{soa4_a1} + 0.0000 \cdot \text{soa5_a2} + 0.0002 \cdot \text{soa5_a1}; 1.e9$
bc_a01	$0.1216 \cdot \text{bc_a1} + 0.1216 \cdot \text{bc_a4}; 1.e9$
bc_a02	$0.7618 \cdot \text{bc_a1} + 0.7618 \cdot \text{bc_a4}; 1.e9$
bc_a03	$0.1164 \cdot \text{bc_a1} + 0.1164 \cdot \text{bc_a4}; 1.e9$
bc_a04	$0.0002 \cdot \text{bc_a1} + 0.0002 \cdot \text{bc_a4}; 1.e9$
so4_a01	$0.9886 \cdot \text{so4_a2} + 0.1216 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
so4_a02	$0.0114 \cdot \text{so4_a2} + 0.7618 \cdot \text{so4_a1} + 0.0002 \cdot \text{so4_a3}; 1.e9$
so4_a03	$0.0000 \cdot \text{so4_a2} + 0.1164 \cdot \text{so4_a1} + 0.0995 \cdot \text{so4_a3}; 1.e9$
so4_a04	$0.0000 \cdot \text{so4_a2} + 0.0002 \cdot \text{so4_a1} + 0.9003 \cdot \text{so4_a3}; 1.e9$
nh4_a01	$0.1856 \cdot \text{so4_a2} + 0.0050 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
nh4_a02	$0.0021 \cdot \text{so4_a2} + 0.0930 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
nh4_a03	$0.0000 \cdot \text{so4_a2} + 0.0203 \cdot \text{so4_a1} + 0.0186 \cdot \text{so4_a3}; 1.e9$
nh4_a04	$0.0000 \cdot \text{so4_a2} + 0.0000 \cdot \text{so4_a1} + 0.1690 \cdot \text{so4_a3}; 1.e9$
no3_a01	$0.0000 \cdot \text{so4_a2} + 0.0000 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
no3_a02	$0.0000 \cdot \text{so4_a2} + 0.0000 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
no3_a03	$0.0000 \cdot \text{so4_a2} + 0.0000 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
no3_a04	$0.0000 \cdot \text{so4_a2} + 0.0000 \cdot \text{so4_a1} + 0.0000 \cdot \text{so4_a3}; 1.e9$
na_a01	$0.3889 \cdot \text{ncl_a2} + 0.0479 \cdot \text{ncl_a1} + 0.0000 \cdot \text{ncl_a3}; 1.e9$
na_a02	$0.0045 \cdot \text{ncl_a2} + 0.2997 \cdot \text{ncl_a1} + 0.0000 \cdot \text{ncl_a3}; 1.e9$
na_a03	$0.0000 \cdot \text{ncl_a2} + 0.0458 \cdot \text{ncl_a1} + 0.0391 \cdot \text{ncl_a3}; 1.e9$
na_a04	$0.0000 \cdot \text{ncl_a2} + 0.0000 \cdot \text{ncl_a1} + 0.3542 \cdot \text{ncl_a3}; 1.e9$
cl_a01	$0.5996 \cdot \text{ncl_a2} + 0.0737 \cdot \text{ncl_a1} + 0.0000 \cdot \text{ncl_a3}; 1.e9$
cl_a02	$0.0068 \cdot \text{ncl_a2} + 0.4621 \cdot \text{ncl_a1} + 0.0000 \cdot \text{ncl_a3}; 1.e9$
cl_a03	$0.0000 \cdot \text{ncl_a2} + 0.0709 \cdot \text{ncl_a1} + 0.0604 \cdot \text{ncl_a3}; 1.e9$
cl_a04	$0.0000 \cdot \text{ncl_a2} + 0.0001 \cdot \text{ncl_a1} + 0.5462 \cdot \text{ncl_a3}; 1.e9$
oin_a01	$0.9886 \cdot \text{dst_a2} + 0.1216 \cdot \text{dst_a1} + 0.0000 \cdot \text{dst_a3}; 1.e9$

oin_a02	0.0114*dst_a2+0.7618*dst_a1+0.0002*dst_a3;1.e9
oin_a03	0.0000*dst_a2+0.1164*dst_a1+0.0995*dst_a3;1.e9
oin_a04	0.0000*dst_a2+0.0002*dst_a1+0.9003*dst_a3;1.e9
num_a01	0.9996*num_a2+0.7135*num_a1+0.0000*num_a3;1.0
num_a02	0.0004*num_a2+0.2847*num_a1+0.0239*num_a3;1.0
num_a03	0.0000*num_a2+0.0016*num_a1+0.6258*num_a3;1.0
num_a04	0.0000*num_a2+0.0000*num_a1+0.3501*num_a3;1.0

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