Supplement of Geosci. Model Dev., 17, 8495–8519, 2024
https://doi.org/10.5194/gmd-17-8495-2024-supplement

Geoscientific Model Development
Open Access
EGU

Supplement of

NAQPMS-PDAF v2.0: a novel hybrid nonlinear data assimilation system for improved simulation of $PM_{2.5}$ chemical components

Hongyi Li et al.

Correspondence to: Ting Yang (tingyang@mail.iap.ac.cn)

Contents of this file

Sect. S1: site classification methodology

This study uses the K-means clustering method to partition the DA and VE sites. The procedure is as follows. Firstly, k initial cluster centers are randomly chosen, and then each observation site is assigned to the cluster closest to its nearest cluster center. After that, the center of each cluster is recalculated, and these two steps are repeated until the cluster centers no longer change or the maximum number of iterations is reached. The number of clusters is determined using the elbow method, which involves plotting the relationship between the number of clusters and the sum of squared errors (SSE) of the clustering results (Fig. S1a). The appropriate number of clusters is identified at the inflection point where the rate of decrease in SSE slows down (14 clusters are identified in this study). The final site division result is shown in Fig. S1b.

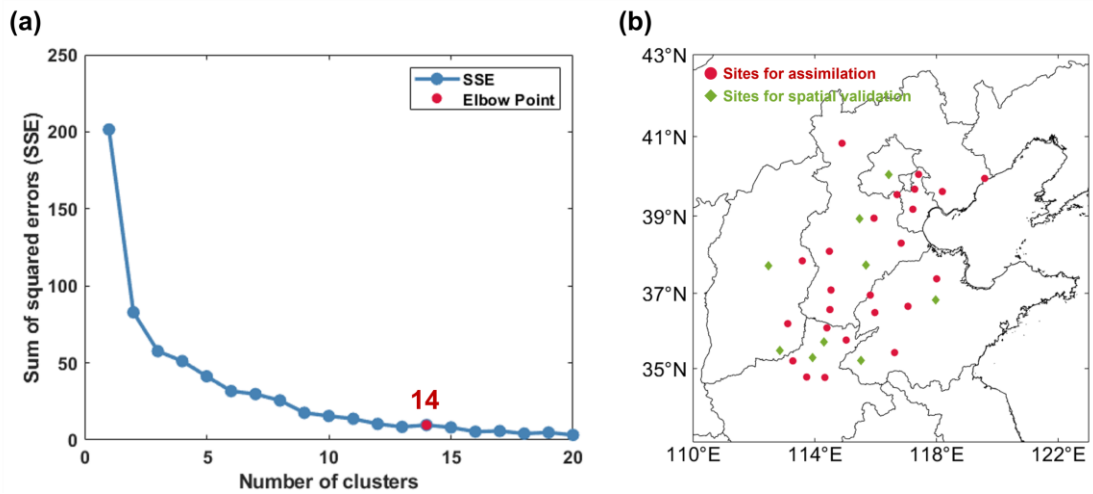

Figure S1: The relationship between the number of clusters and the sum of squared errors (a), the site division result (b), the red sites for assimilation, and the green sites for spatial independent validation.

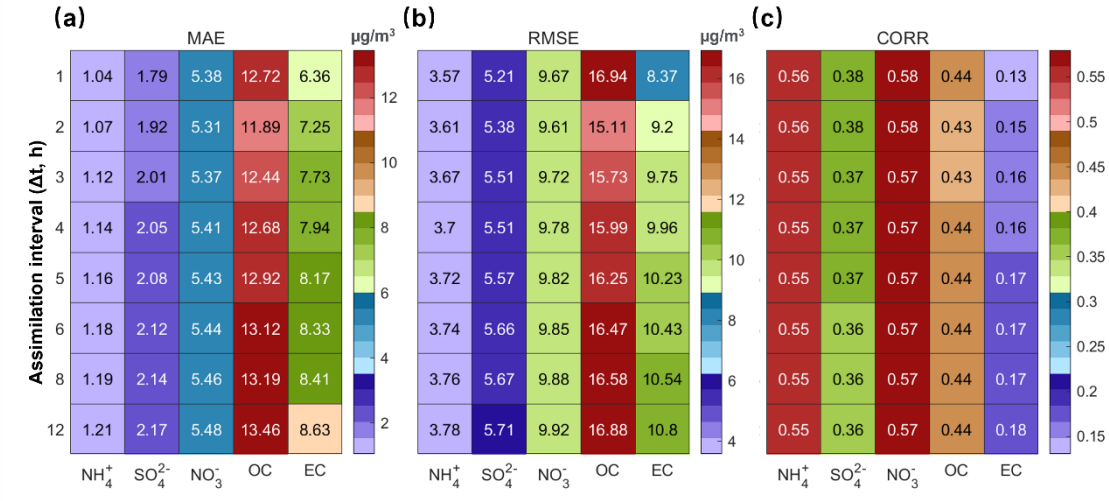

Figure S2: Assessment of assimilation interval dependency based on mean absolute error (MAE) (a), root

mean square error (RMSE) (b), correlation coefficient (CORR) (c) at forecast step.

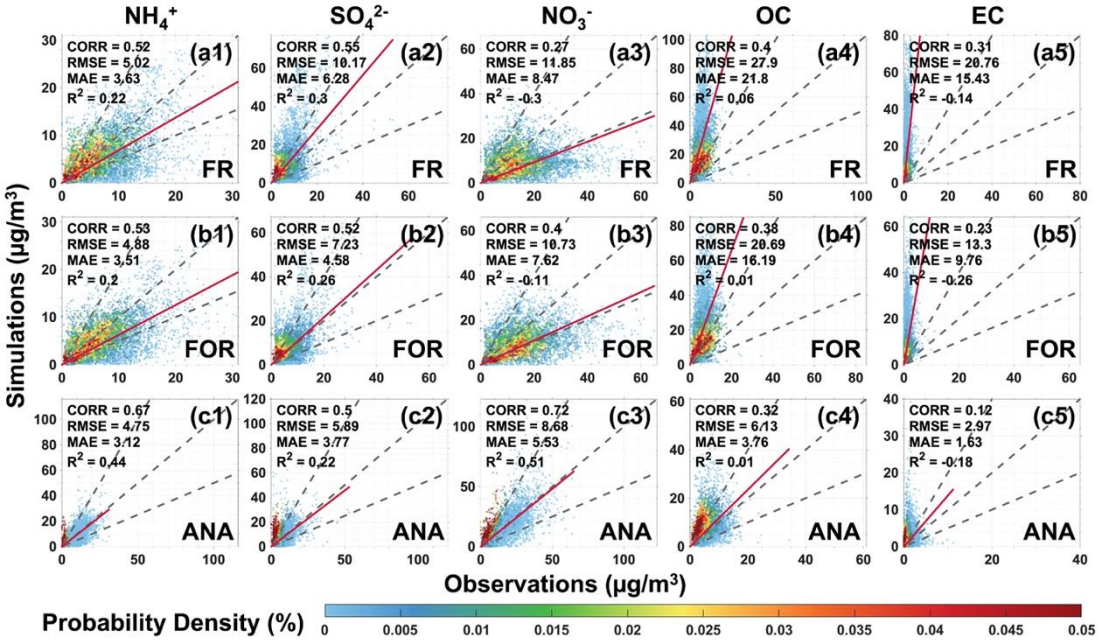

Figure S3: Scatterplots of the VE site simulations versus the VE site observations with probability density for the free-running (FR) field (a1-a5), forecast (FOR) field (b1-b5), and analysis (ANA) field (c1-c5). The stippled gray lines represent the 2:1, 1:1, and 1:2 lines, and the solid red line represents the fitting regression line.

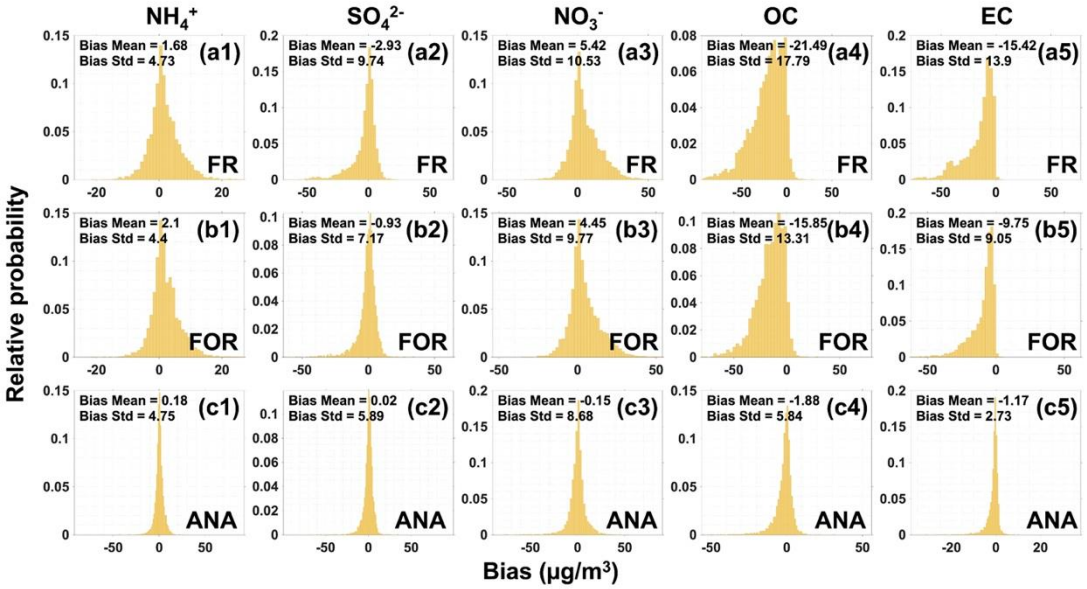

Figure S4: Probability distributions of bias between VE site observations and VE site simulations for the free-running (FR) field (a1-a5), forecast (FOR) field (b1-b5), and analysis (ANA) field (c1-c5).

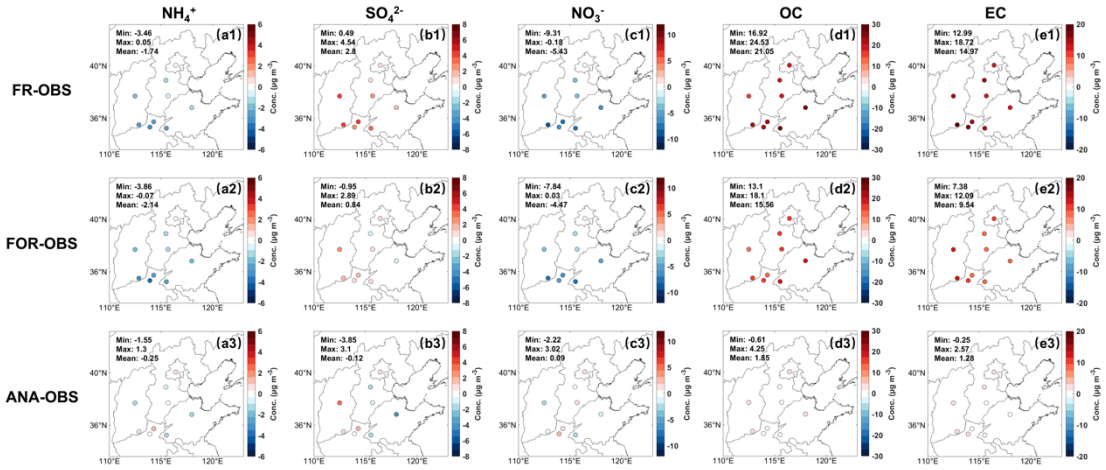

Figure S5: Spatial distribution of VE site bias for five $PM_{2.5}$ chemical components from the observation (OBS) for the free-running (FR) field (a1-e1), forecast (FOR) field (a2-e2), and analysis (ANA) field (a3-e3).

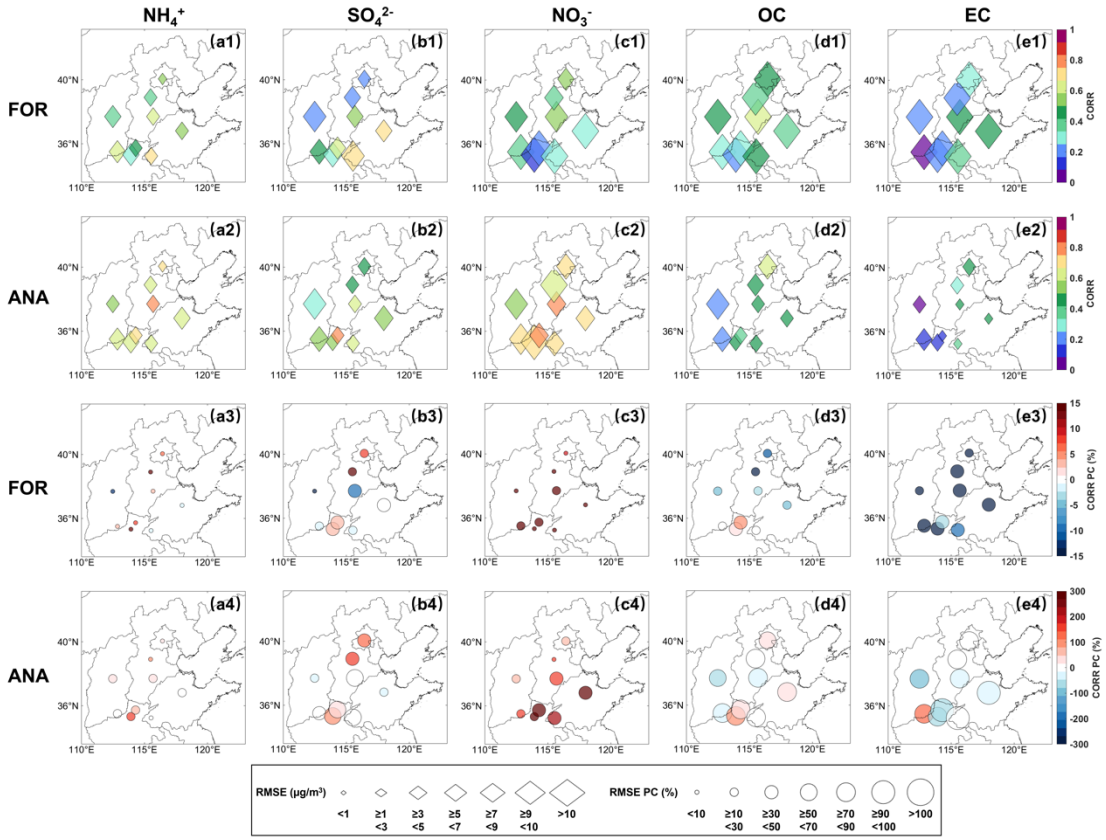

Figure S6: Spatial distribution of VE site statistical indicators for five $PM_{2.5}$ chemical components. Panels (a1-e1) represent the values of RMSE and CORR for the forecast (FOR) field, (a2-e2) same as (a1-e1) but for the analysis (ANA) field, (a3-e3) represent the improvement in RMSE and CORR for the forecast (FOR) field, and (a4-e4) are the same as (a3-e3) but for the analysis (ANA) field. The size represents the value of RMSE in (a1-e2) and the improvement percentage compared to non-assimilation in (a3-e4).

Table S1: The physicochemical process parameterization schemes in NAQPMS.

Process	Scheme	Feature	Reference
Advection process	High precision positive definite mass conserving difference scheme	Fixed-grid size, mass conserving, stability, computational accuracy	(Walcek and Aleksic, 1998)
Eddy diffusion	Gradient transport theory	High precision, local K theory scheme	(Byun and Dennis, 1995)
Dry deposition	Resistance model	Consideration of gravity deposition for aerosol	(Wesely, 2007; Slinn and Slinn, 1980)
Wet scavenging	$W_{ash} = W_a C_i$	Calculation of gas and particulate matter	\
Gas-phase chemistry	Carbon Bond Mechanism version Z (CBM-Z)	71 species and 134 reactions, lumped structure, carbon number conservation, computational efficiency	(Zaveri and Peters, 1999)
Aqueous chemistry	Regional Acid Deposition Model version 2 (RADM2)	63 species and 157 reactions, high vertical coverage range	(Stockwell et al., 1990)
Aerosol physicochemical process	Thermodynamic equilibrium model (ISORROPIA), Secondary Organic Aerosol Production (SOAP) model	Calculation of inorganic aerosol fractions and secondary organic aerosol, respectively	(Nenes et al., 1998; Strader et al., 1999)
Heterogeneous reactions	The first-order rate constant (k_{het})	28 reactions, simulation of the mixing of aerosols with anthropogenic gases	(Li et al., 2012)
Other processes	Radiative transfer model, online emission calculation	Simulation of the mixing of aerosols with gases, calculation of dimethyl sulfide, sea salt, and dust	(Li et al., 2011; Li et al., 2018; Lana et al., 2011; Athanasopoulou et al., 2008; Wang et al., 2000)

Table.S2. Parameterization schemes in WRF.

Physical processes	Parameterization schemes	Reference
Cloud microphysics process	Lin scheme	(Lin et al., 1983)
Longwave radiation	Rapid Radiative Transfer Model (RRTM)	(Mlawer et al., 1997)
Shortwave radiation	Goddard scheme	(Chou and Suarez, 1999)
Planetary boundary layer process	Yonsei University (YSU) scheme	(Hong et al., 2006)
Cumulus convection process	Kain-Fritsch scheme	(Kain and Fritsch, 1990)
Land surface processes	Noah Land Surface Model (LSM)	(Niu et al., 2011)
Surface layer scheme	MM5 similarity theory scheme	(Hari Prasad et al., 2016)

Table S3: The statistical indicators (CORR and RMSE) in the free-running field (FR), forecast field (FOR), and analysis field (ANA) at three representative DA sites and three representative VE sites.

	Stations	FR		FOR		ANA	
		CORR	RMSE (μg/m^3)	CORR	RMSE (μg/m^3)	CORR	RMSE (μg/m^3)
DA	Tianjin	0.66	52.26	0.67	38.78	0.94	22.98
	Langfang	0.61	50.73	0.63	35.15	0.94	14.08
	Jiaozuo	0.48	67.86	0.52	41.19	0.91	11.45
VE	Beijing	0.48	58.53	0.46	49.10	0.75	19.57
	Zibo	0.66	73.87	0.64	51.61	0.75	19.80
	Xinxiang	0.49	66.21	0.51	41.18	0.71	22.10

Reference

Athanasopoulou, E., Tombrou, M., Pandis, S., and Russell, A.: The role of sea-salt emissions and heterogeneous chemistry in the air quality of polluted coastal areas, Atmos. Chem. Phys., 8, 5755-5769, https://doi.org/10.5194/acp-8-5755-2008, 2008.

Byun, D. W. and Dennis, R.: Design artifacts in eulerian air quality models: Evaluation of the effects of layer thickness and vertical profile correction on surface ozone concentrations, Atmos. Environ., 29, 105-126, https://doi.org/10.1016/1352-2310(94)00225-A, 1995.

Chou, M.-D. and Suarez, M. J.: A solar radiation parameterization for atmospheric studies, (No. NASA/TM-1999-104606/VOL15). Greenbelt, MD: NASA Goddard Space Flight Center, 1999.

Hari Prasad, K. B. R. R., Venkata Srinivas, C., Venkateswara Naidu, C., Baskaran, R., and Venkatraman, B.: Assessment of surface layer parameterizations in ARW using micro-meteorological observations from a tropical station, Meteorol. Appl., 23, 191-208, https://doi.org/10.1002/met.1545, 2016.

Hong, S.-Y., Noh, Y., and Dudhia, J.: A new vertical diffusion package with an explicit treatment of entrainment processes, Mon. Weather Rev., 134, 2318-2341, https://doi.org/10.1175/MWR3199.1, 2006.

Kain, J. S. and Fritsch, J. M.: A One-Dimensional Entraining/Detraining Plume Model and Its Application in Convective Parameterization, J. Atmos. Sci., 47, 2784-2802, https://doi.org/10.1175/1520-0469(1990)047<2784:AODEPM>2.0.CO;2, 1990.

Lana, A., Bell, T., Simó, R., Vallina, S., Ballabrera-Poy, J., Kettle, A., Dachs, J., Bopp, L., Saltzman, E., and Stefels, J.: An updated climatology of surface dimethlysulfide concentrations and emission fluxes in the global ocean, Global Biogeochem. Cy., 25, https://doi.org/10.1029/2010GB003850, 2011.

Li, J., Wang, Z., Zhuang, G., Luo, G., Sun, Y., and Wang, Q.: Mixing of Asian mineral dust with anthropogenic pollutants over East Asia: a model case study of a super-duststorm in March 2010, Atmos. Chem. Phys., 12, 7591-7607, https://doi.org/10.5194/acp-12-7591-2012, 2012.

Li, J., Chen, X., Wang, Z., Du, H., Yang, W., Sun, Y., Hu, B., Li, J., Wang, W., and Wang, T.: Radiative and heterogeneous chemical effects of aerosols on ozone and inorganic aerosols over East Asia, Sci. Total Environ., 622-623, 1327-1342, https://doi.org/10.1016/j.scitotenv.2017.12.041, 2018.

Li, J., Wang, Z., Wang, X., Yamaji, K., Takigawa, M., Kanaya, Y., Pochanart, P., Liu, Y., Irie, H., and Hu, B.: Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China, Atmos. Environ., 45, 1817-1829, https://doi.org/10.1016/j.atmosenv.2011.01.016,

2011.

Lin, Y.-L., Farley, R. D., and Orville, H. D.: Bulk Parameterization of the Snow Field in a Cloud Model, J. Appl. Meteorol. Clim., 22, 1065-1092, https://doi.org/10.1175/1520-0450(1983)022<1065:BPOTSF>2.0.CO;2, 1983.

Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, J. Geophys. Res.-Atmos., 102, 16663-16682, https://doi.org/10.1029/97JD00237, 1997.

Nenes, A., Pandis, S. N., and Pilinis, C.: ISORROPIA: A new thermodynamic equilibrium model for multiphase multicomponent inorganic aerosols, Aquat. Geochem., 4, 123-152, https://doi.org/10.1023/A:1009604003981, 1998.

Niu, G. Y., Yang, Z. L., Mitchell, K. E., Chen, F., Ek, M. B., Barlage, M., Kumar, A., Manning, K., Niyogi, D., and Rosero, E. J. J. o. G. R. A.: The community Noah land surface model with multiparameterization options (Noah-MP): 1. Model description and evaluation with local-scale measurements, J. Geophys. Res.-Atmos., 116, https://doi.org/10.1029/2010JD015139, 2011.

Slinn, S. A. and Slinn, W. G. N.: Predictions for particle deposition on natural waters, Atmos. Environ. (1967), 14, 1013-1016, https://doi.org/10.1016/0004-6981(80)90032-3, 1980.

Stockwell, W. R., Middleton, P., Chang, J. S., and Tang, X.: The second generation regional acid deposition model chemical mechanism for regional air quality modeling, J. Geophys. Res.-Atmos., 95, 16343-16367, https://doi.org/10.1029/JD095iD10p16343, 1990.

Strader, R., Lurmann, F., and Pandis, S. N.: Evaluation of secondary organic aerosol formation in winter, Atmos. Environ., 33, 4849-4863, https://doi.org/10.1016/S1352-2310(99)00310-6, 1999.

Walcek, C. J. and Aleksic, N. M.: A simple but accurate mass conservative, peak-preserving, mixing ratio bounded advection algorithm with FORTRAN code, Atmos. Environ., 32, 3863-3880, https://doi.org/10.1016/S1352-2310(98)00099-5, 1998.

Wang, Z., Ueda, H., and Huang, M.: A deflation module for use in modeling long-range transport of yellow sand over East Asia, J. Geophys. Res.-Atmos., 105, 26947-26959, https://doi.org/10.1029/2000JD900370, 2000.

Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 41, 52-63, https://doi.org/10.1016/j.atmosenv.2007.10.058, 2007.

Zaveri, R. A. and Peters, L. K.: A new lumped structure photochemical mechanism for large-scale

applications, J. Geophys. Res.-Atmos., 104, 30387-30415, https://doi.org/10.1029/1999JD900876, 1999.