Development of a Regional multi-Air Pollutant Assimilation System and its applications in emission estimates in China

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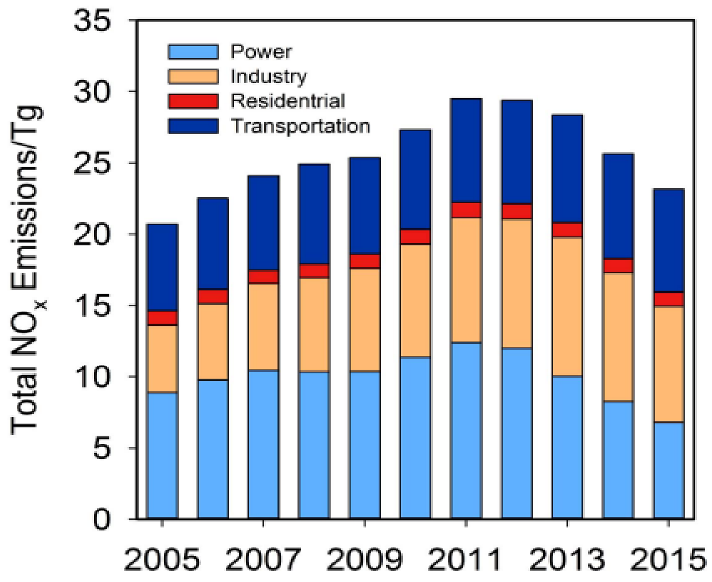
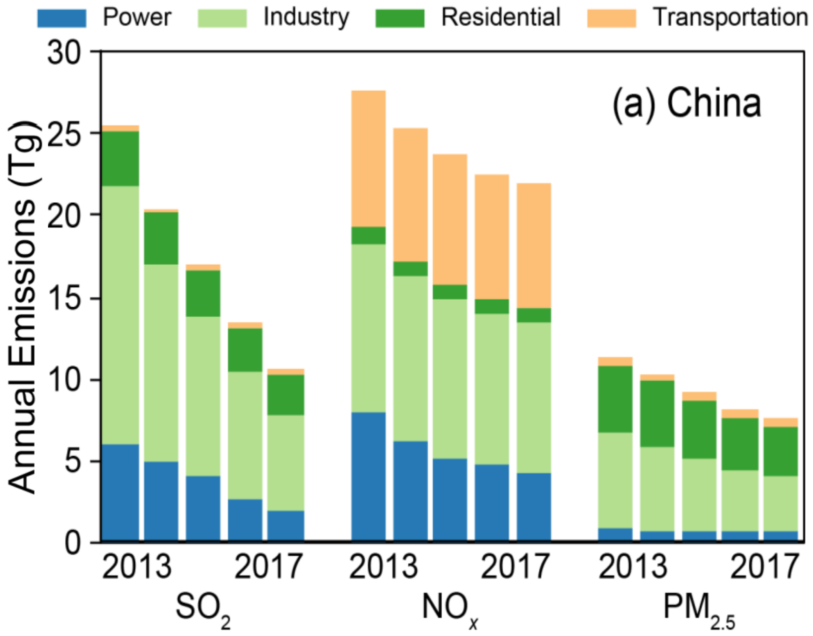
1. Background

- ✓ China's air pollutant emissions have **large inter-annual changes**, but the release of emission inventories usually **lags 1-3 years**, leading the latest inventory cannot represent the current actual emission situation.
- ✓ The bottom-up emission inventories still have **large uncertainties**.
- ✓ Making the emission inventories unable to well support the predictions and management of air quality in China



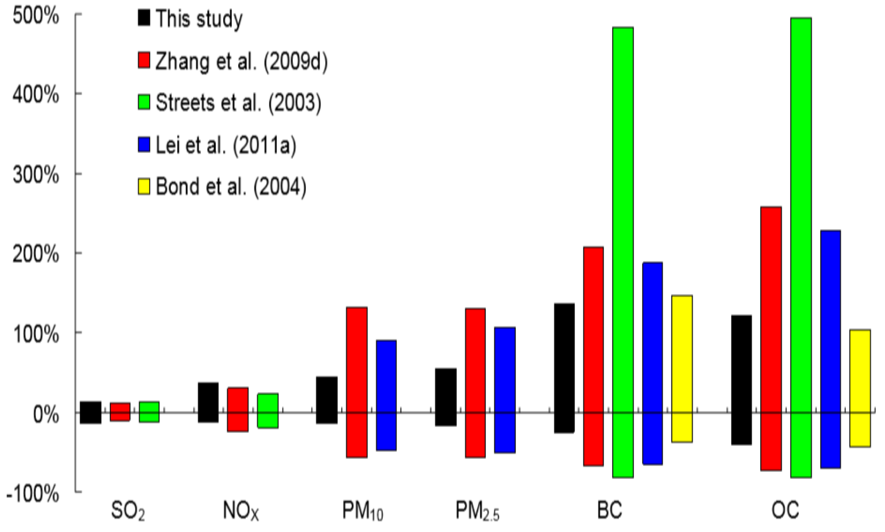
Total anthropogenic NOx emissions by sector for China during 2005–2015

Liu et al. 2016@ERL



Deceases of SO_2, NO_x, and $PM_{2.5}$ emissions from 2013 to 2017 in China due to clean air actions

Zhang et al., 2019@PNAS



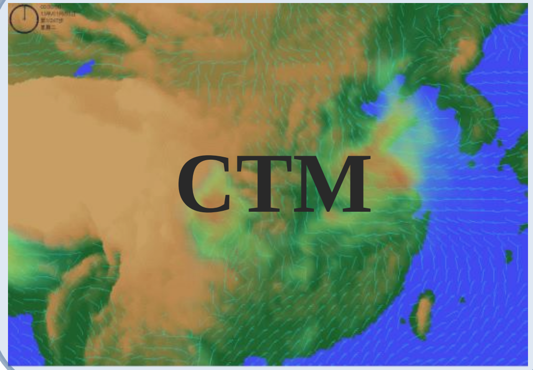
Emission uncertainties for different species in China from different studies

Zhao et al., 2011, ACP

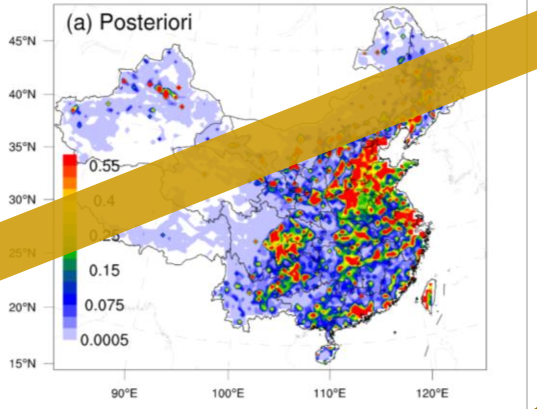
1. Background

Top-down atmospheric inversion uses bottom-up emission inventories as a priori, and spatially distributed observations as constraints, which is one of the ways to improve the emission estimates and update the emissions timely.

Satellite and aircraft observations



Posterior emission

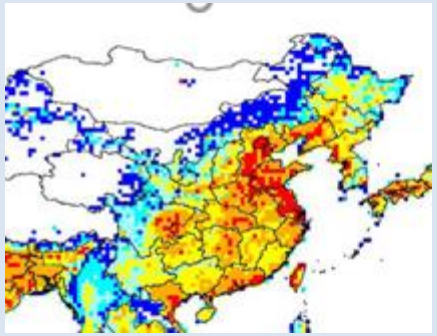


Top-down inversion

Emission inventory

industry

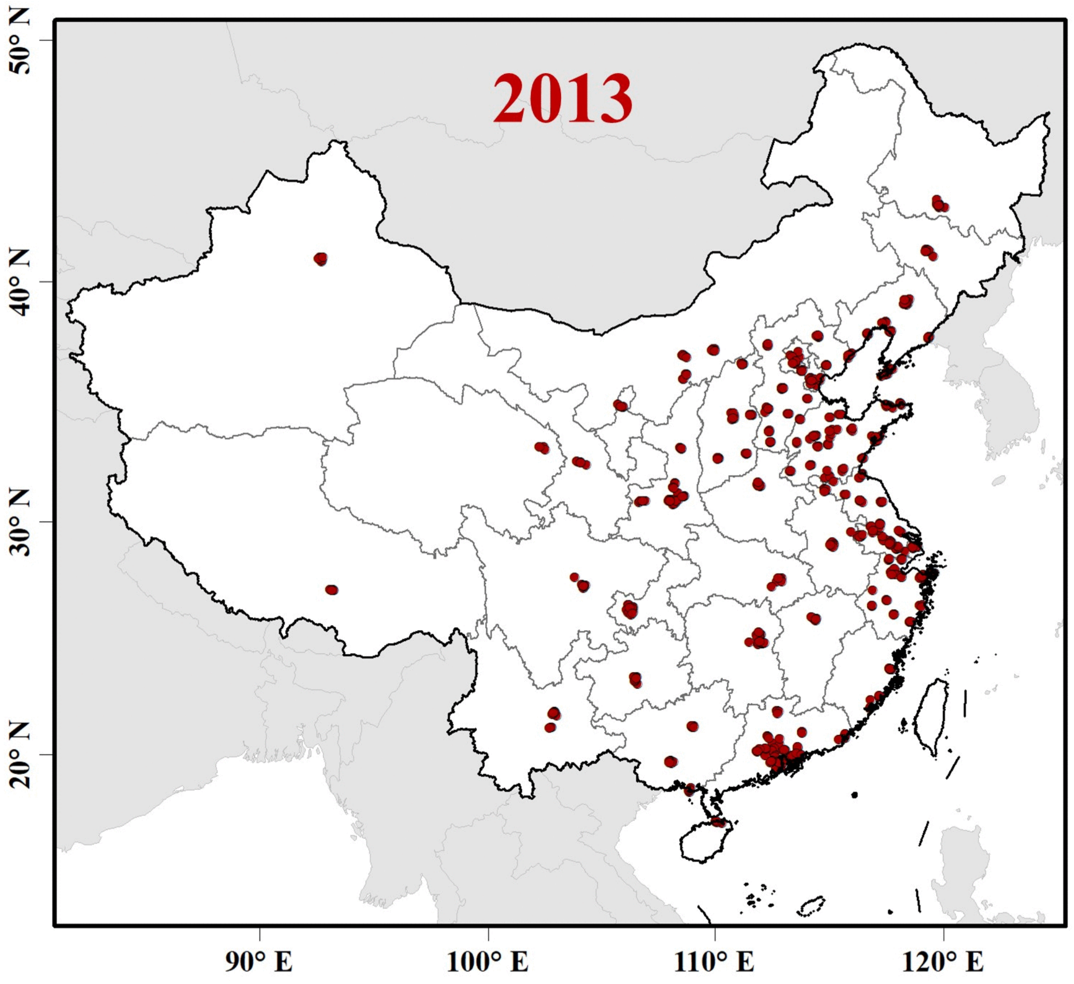
transport



Surface measurement

1. Background

- Since 2013, China has gradually established a nationwide air pollution monitoring network (> 1600 national control sites). The monitoring species include SO_2, NO_2, CO, O_3, $PM_{2.5}$, and PM_{10}. These measurements are released hourly by China National Environmental Monitoring Centre
- Advantages of emission inversion based on surface observations
  - ✓ High precision, not affected by the weather
  - ✓ Mostly located in urban, can quickly sense emission changes
  - ✓ hourly observations
- **Our goal is to establish an operational emission inversion system based on ground observations, to support air quality forecasting and management.**

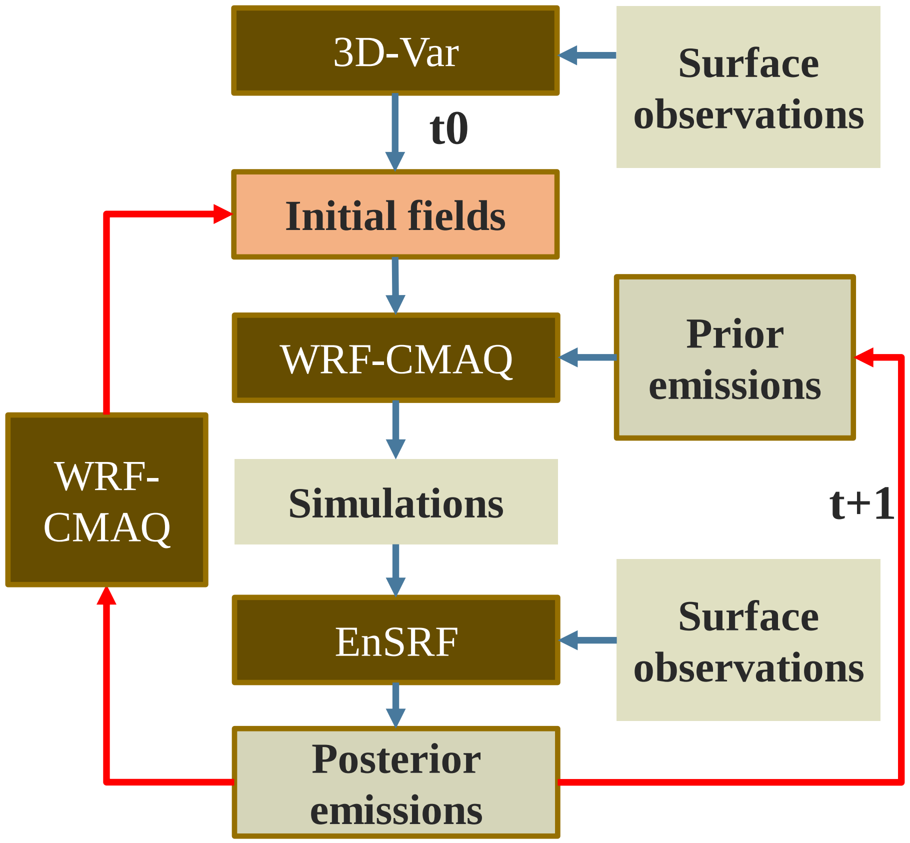


Locations of national control monitoring sites

2. System description

A Regional multi-Air Pollutant Assimilation System (RAPAS v1.0)

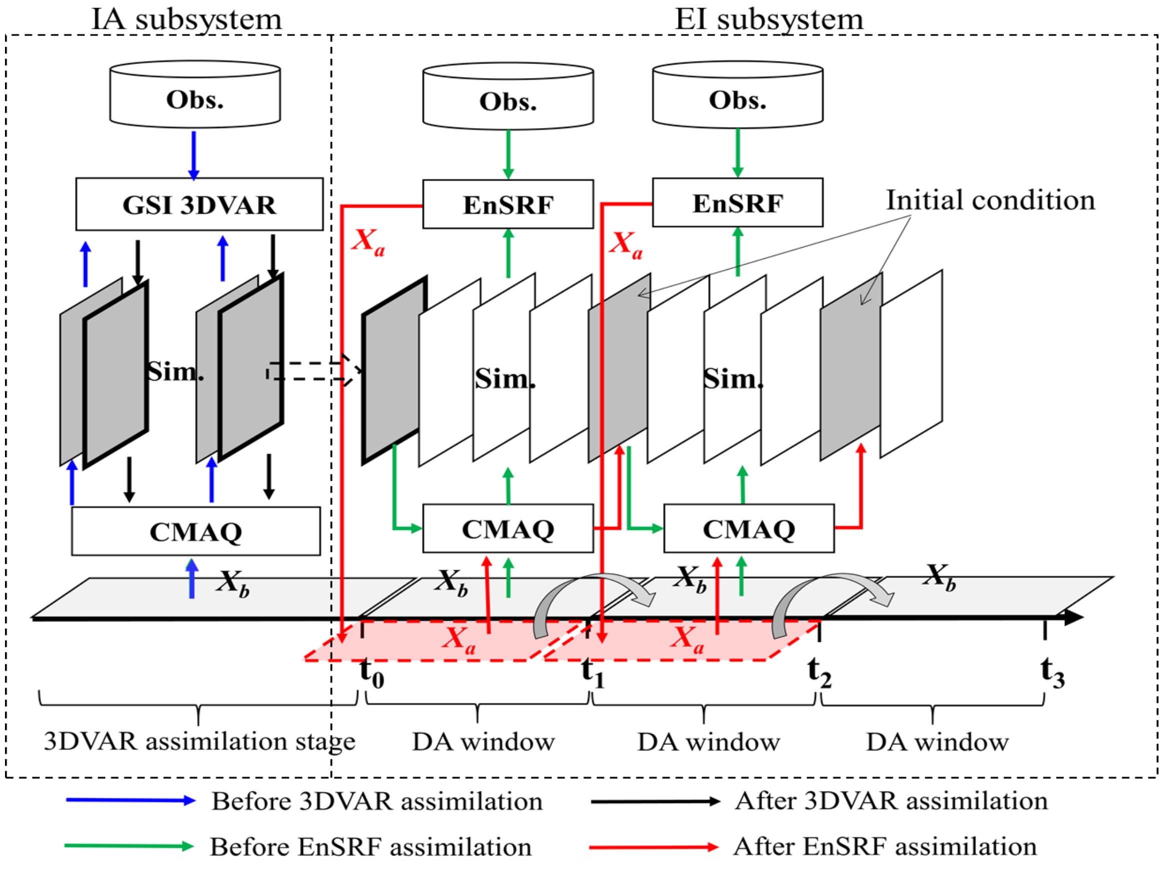
- ✓ First, the prior emissions in each grid are optimized through data assimilation,
- ✓ Second, the initial field of the next window is optimized using the posterior emissions through forward simulation,
- ✓ Meanwhile, the posterior emissions are transferred to the next window as its prior emissions.



Flow chart of RAPAS v1.0

- CTM: WRFv4.0 + CMAQv5.0.2
- Initial field DA: 3D-Var
- Emission inversion: EnSRF
- The emissions of SO_2, NO_x, CO, primary $PM_{2.5}$ and coarse PM_{10} are inferred simultaneously; point and area emissions are estimated separately

2. System description



Detailed operation process

- The system is divided into IA and EI subsystems, IA is to provide a well chemical ICs for the following EI, and EI is run cyclically to optimize the emissions
- **Number of ensembles**: 40
- **DA window**: 1 day, due to the complexity of hourly emissions, it is very difficult to simulate hourly concentrations that can match the observations well
- **Localization scale**: set according to the DA window, mean wind speed, and lifetime of different species

Species	CO	SO_2	NO_2	$PM_{2.5}$	PMC
Lifetime	~2 m	> 1 d	~10 h	> 1 d	< 1 d
Localization (km)	**300**	**300**	**150**	**300**	**250**

2. System description

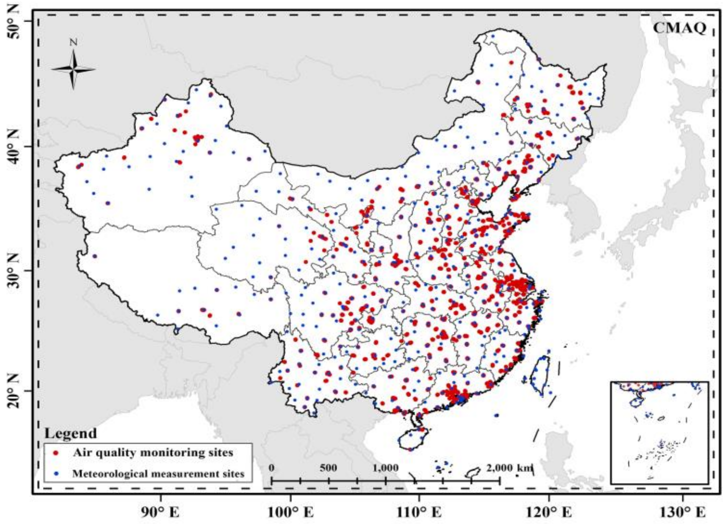
Model domain and main configurations

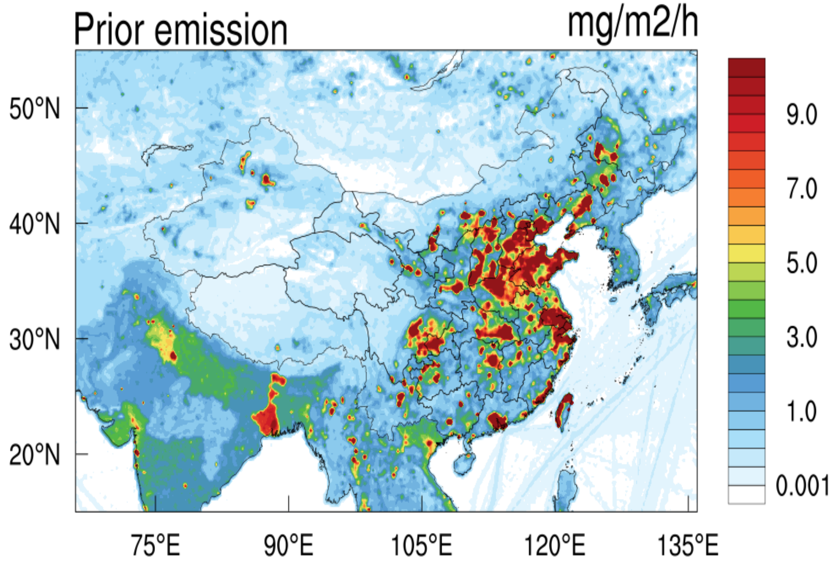
- covers the whole mainland of China, with grid spacing of 36 km
- Vertical layers: WRF 51 sigma levels, and CMAQ 16 layers
- Gas phase and aerosol scheme: CB05tucl + AERO6
- Chemical boundary conditions: idea profile

Prior emissions

- Multi-resolution Emission Inventory for China (MEIC, 0.25°); Mosaic Asian Anthropogenic Emission Inventory for outside China (MIX, 0.25°)
- The uncertainties of SO_2, NO_x, CO, primary $PM_{2.5}$ and coarse PM_{10} emissions are set to 25%, 25%, 30%, 40%, and 40% in each model grid.
- Biogenic emissions are not optimized, biomass burning emissions are not included.

Model domain





2. System description

Observations

- Hourly averaged observations (SO_2, NO_2, CO , $PM_{2.5}$ and PM_{10}) from over 1600 national control air quality stations
- **Super-observation**: A super-observation is generated by averaging all observations located within the same model grid within a DA window.

$$1/r_{new}^2 = \sum_{j=1}^{m} 1/r_j^2$$

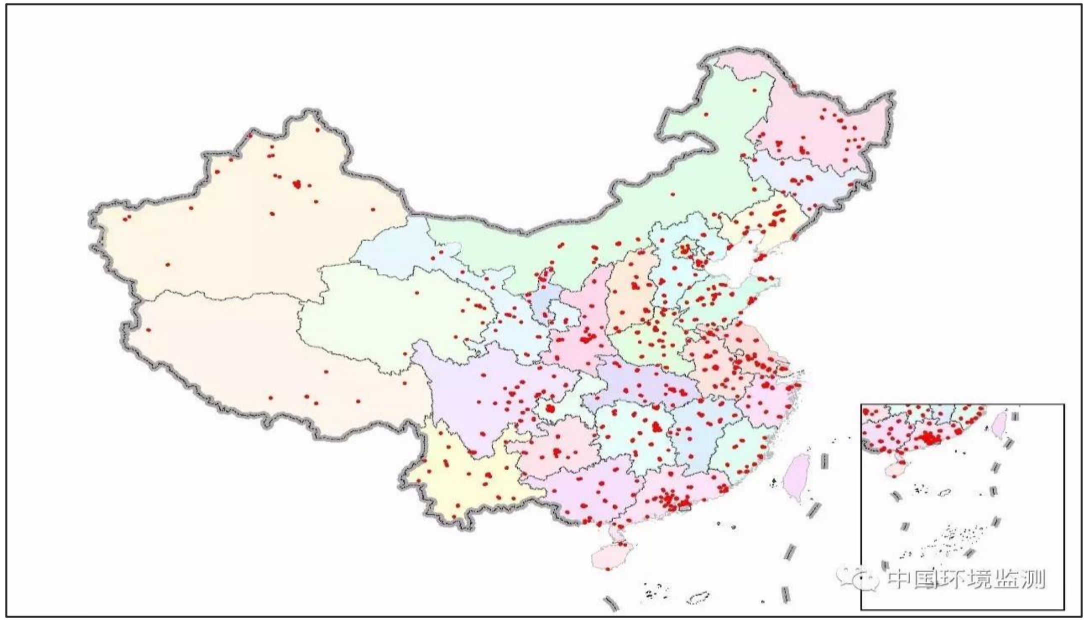
$$y_{new} = \sum_{j=1}^{m} w_j\, y_j / \sum_{j=1}^{m} w_j$$

$$x_{new,i} = \sum_{j=1}^{m} w_j\, x_{ij} / \sum_{j=1}^{m} w_j$$

Observation error

- Measurement error, $\varepsilon_0 = ermax + ermin * \Pi_0$
- Representation error, $\varepsilon_r = \gamma \varepsilon_0 \sqrt{\Delta x / L}$ (γ, tunable parameter; Δx, grid spacing; L, observation scale)
- Total error, $\varepsilon = \sqrt{{\varepsilon_0}^2 + {\varepsilon_r}^2}$

Locations of the surface observations

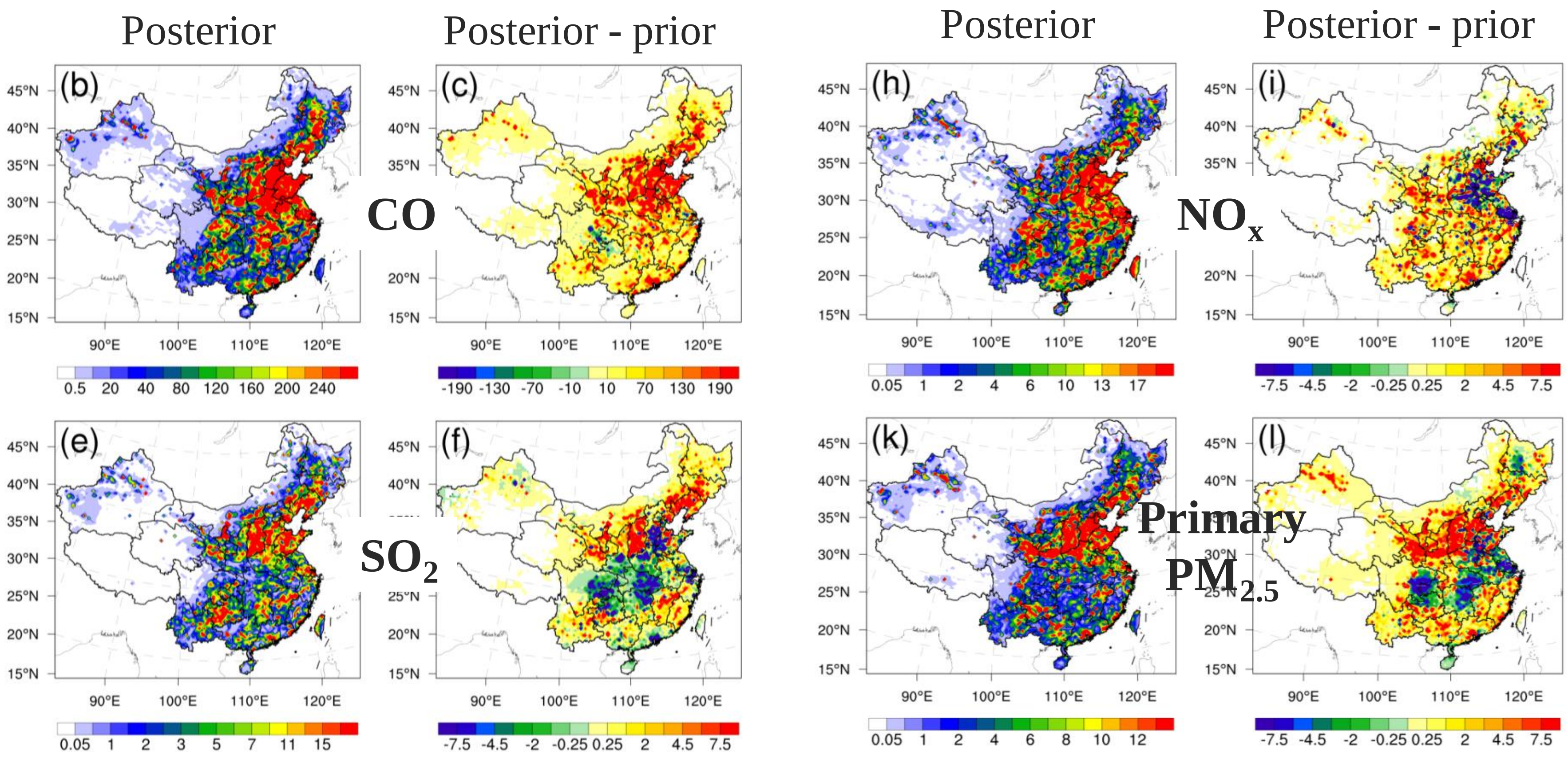


Parameter	CO mg/m^3	SO_2 µg/m^3	NO_2 µg/m^3	O_3 µg/m^3	$PM_{2.5}$ µg/m^3	PMC µg/m^3
value-range check	0.1~12	1~800	1~250	1~250	1~800	1~900
time-continuity check	2.5	160	70	80	180	180
ermax	0.05	1	1	1	1.5	1.5
ermin	0.5%	0.5%	0.5%	0.5%	0.75%	0.75%

2. System evaluations

Emission estimates for December 2016

Prior emission: MEIC 2016; **Obs.**: 1504 national control sites



Compared with prior emission,

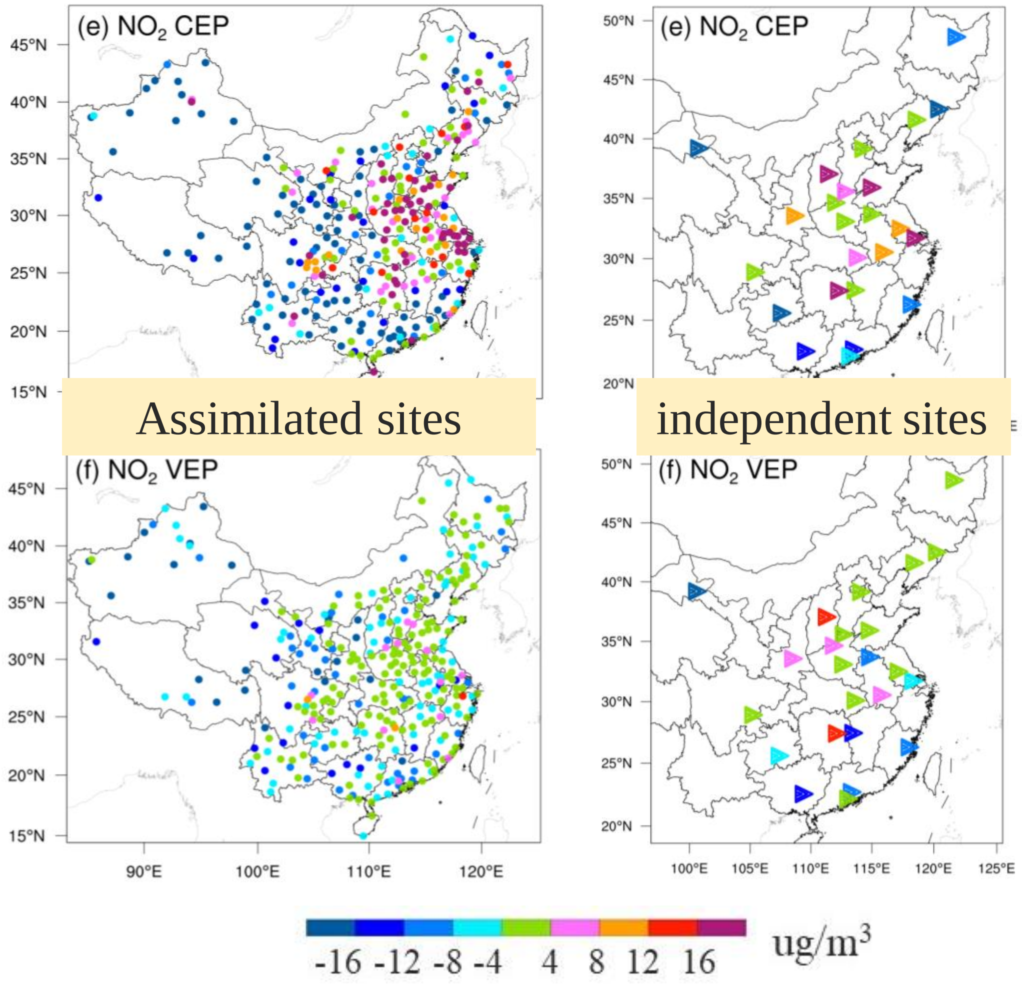
- ✓ The optimized CO emissions are increased across most areas of mainland China, especially in northern China.
- ✓ The inverted NOx emissions are decreased over YRD and north China plain, while in southern, western China, they are increased.
- ✓ For SO_2 and primary $PM_{2.5}$, the inverted emissions are decreased over YRD, part of North China plain, and central China.

The CO, SO_2, NO_x, and primary $PM_{2.5}$ emissions increase by 129%, 20%, 5%, and 95%, respectively.

2. System evaluations

Evaluating the simulated concentrations with observations

- ✓ With prior emission, large negative or positive biases, with posterior emissions, in most sites, the biases are largely reduced.
- ✓ The RMSE decreases by about 40% and 30% against assimilated and independent observations, CORR significantly increased.

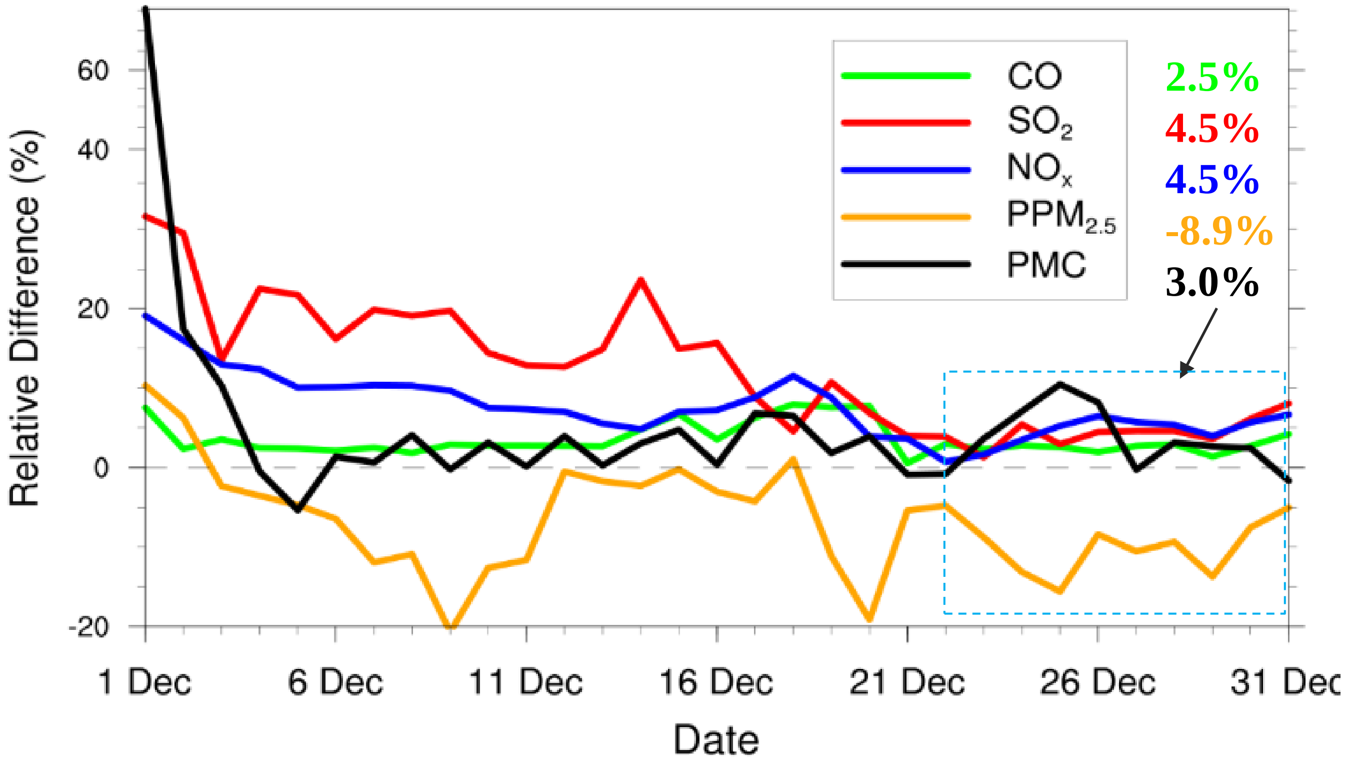


Distribution of mean biases at different sites

Species	Mean Obs.	Mean Sim.		BIAS		RMSE		CORR	
		CEP	VEP	CEP	VEP	CEP	VEP	CEP	VEP
Against assimilated observations									
CO	1.43	0.66	1.36	-0.77	-0.08	1.08	0.56	0.46	0.81
SO_2	32.5	34.4	28.4	1.9	-4.1	42.4	17.7	0.39	0.88
NO_2	43.8	40.8	39.0	-2.9	-4.8	25.0	12.3	0.65	0.88
$PM_{2.5}$	77.0	53.1	70.3	-24.0	-6.7	50.3	29.6	0.64	0.87
PMC	40.5	8.1	37.5	-32.4	-3.1	41.5	24.6	0.25	0.69
Against independent observations									
CO	1.54	0.79	1.52	-0.75	-0.02	1.15	0.72	0.59	0.82
SO_2	40.6	39.2	37.3	-1.3	-3.2	44.3	27.2	0.57	0.87
NO_2	50.2	50.0	47.5	-0.3	-2.7	21.7	15.9	0.73	0.83
$PM_{2.5}$	91.5	64.6	84.1	-26.9	-7.4	64.1	37.2	0.62	0.87
PMC	42.0	9.2	40.4	-32.8	-1.6	39.3	26.6	0.39	0.62

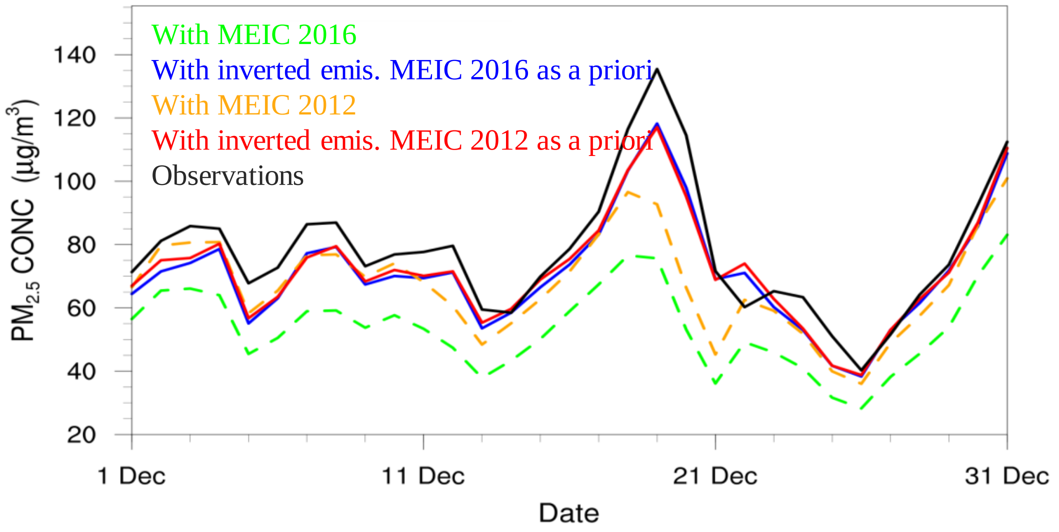
CEP: Simulation experiment using original prior emissions
VEP: simulation experiment using posterior emissions

MEIC 2012 and MEIC 2016 were used as a priori for the emission estimates in December 2016



Relative differences in the inverted CO, SO_2, NO_x, primary $PM_{2.5}$ and coarse PM10 emissions using different prior emissions

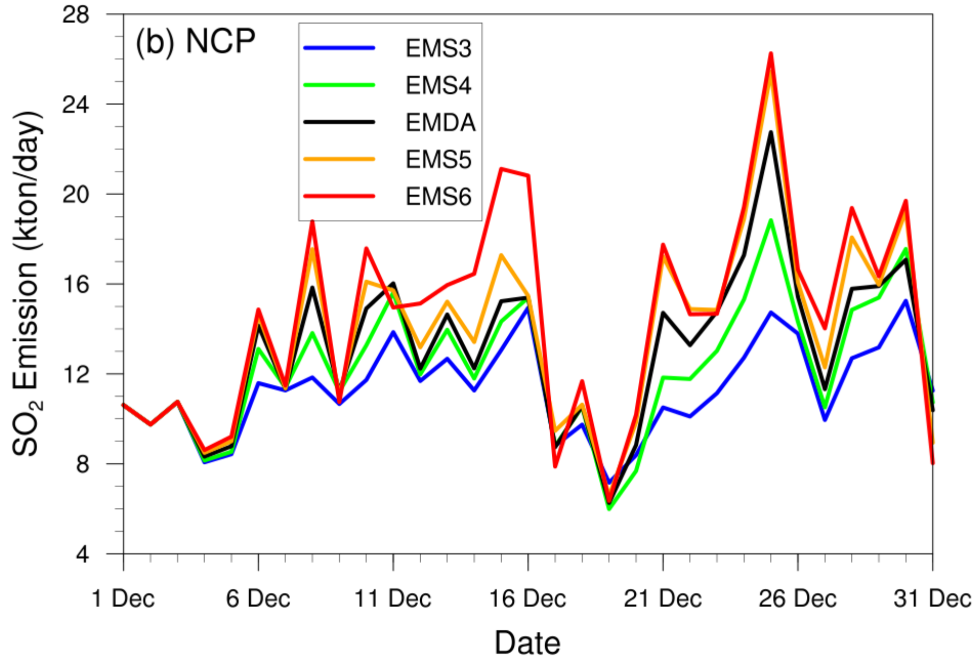
- The differences between the two posterior emissions gradually decrease over time, the quick convergence of PMC is due to large prior uncertainty of 100% used in the first 3 DA windows.
- More SO_2 and NO_x emissions result in more secondary $PM_{2.5}$, leading to less primary $PM_{2.5}$ emission estimated.



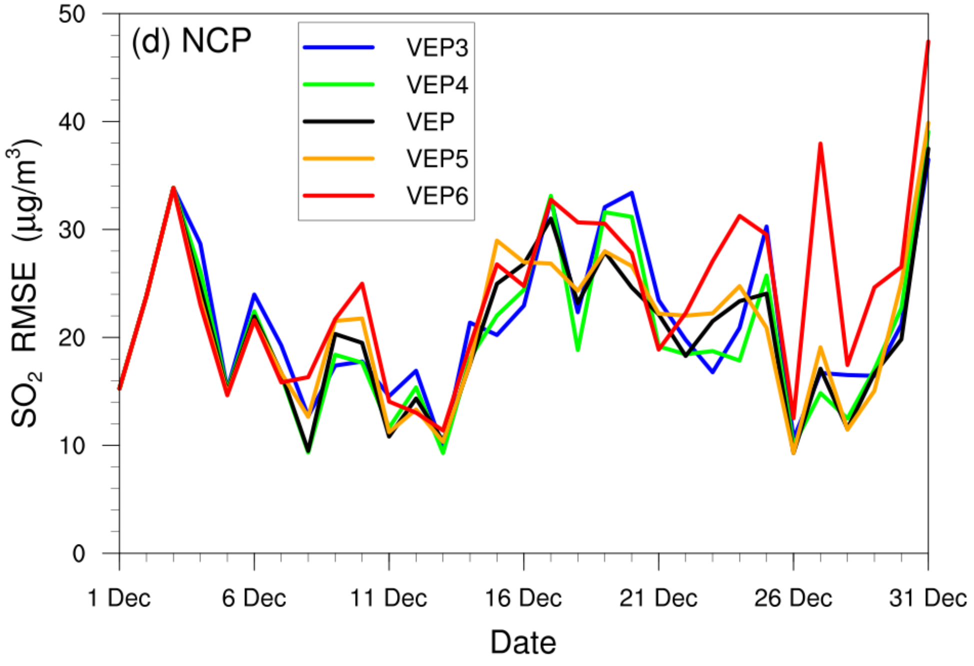
Observed and simulated daily $PM_{2.5}$ concentrations

2. System evaluations Impact of emission uncertainty settings on emission estimates

Inverted daily SO_2 emissions



Daily RMSE of simulated SO_2 conc.



EMS3： -50%

EMS4： -25%

EMDA： default

EMS5： +25%

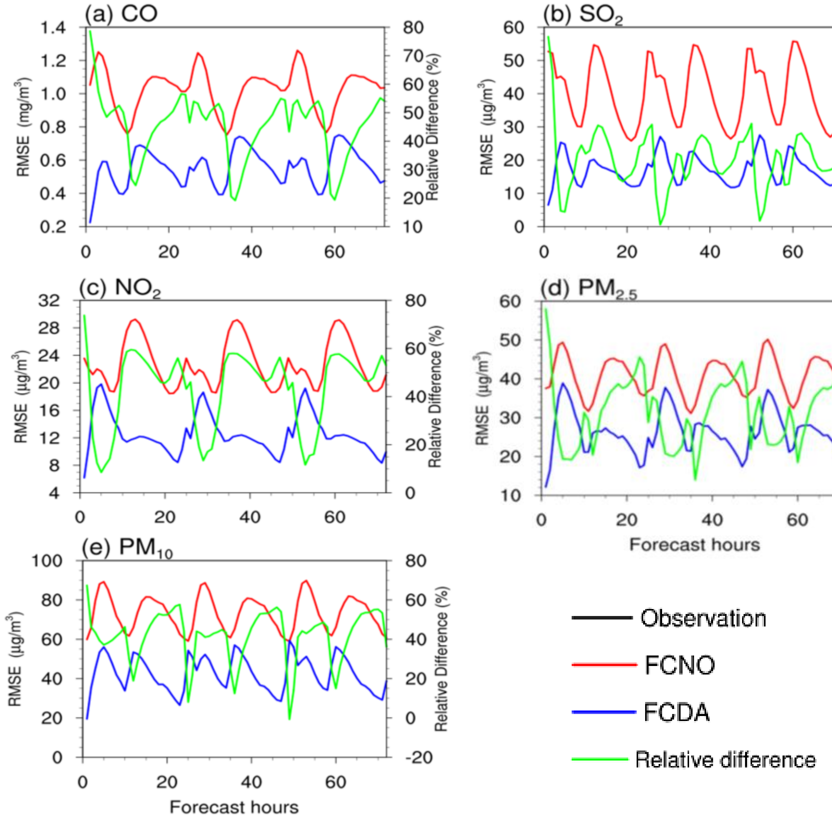
EMS6： +50%

- A larger prior emission uncertainty, results in larger day-to-day variation of inverted emission, and stronger emission.
- When the uncertainty increases or decreases, the RMSE of the simulated concentrations with posterior emissions will increase.

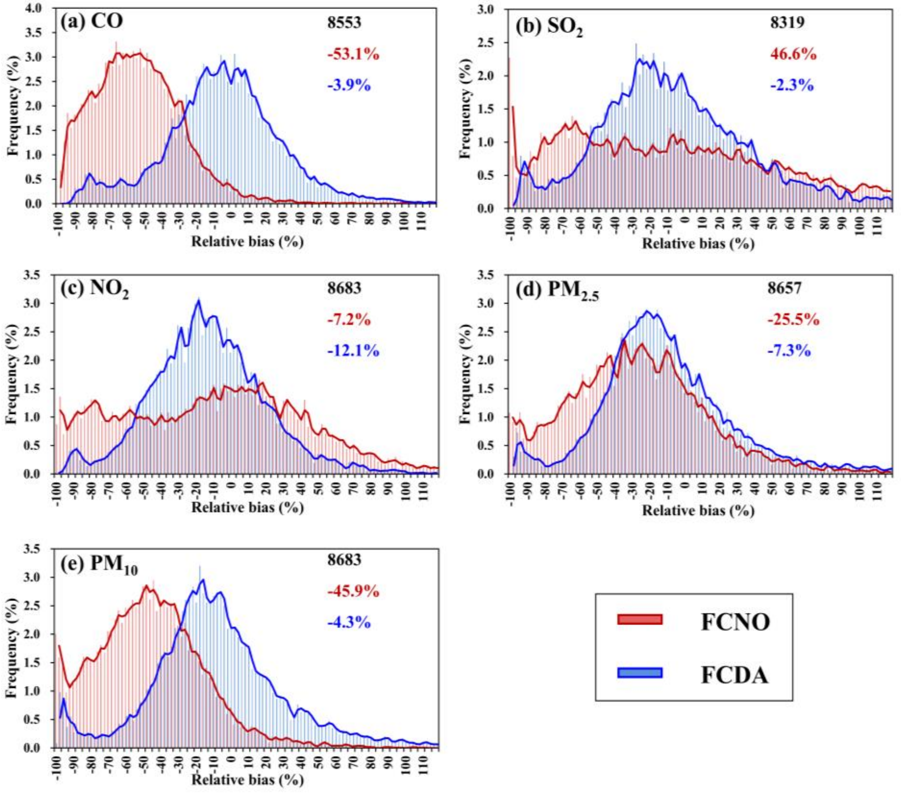
3. Applications

Impact on the regional air quality forecasts in China

The inverted emissions of current day are used in the forecasts for next 72 hours every day



Time series of hourly RMSE

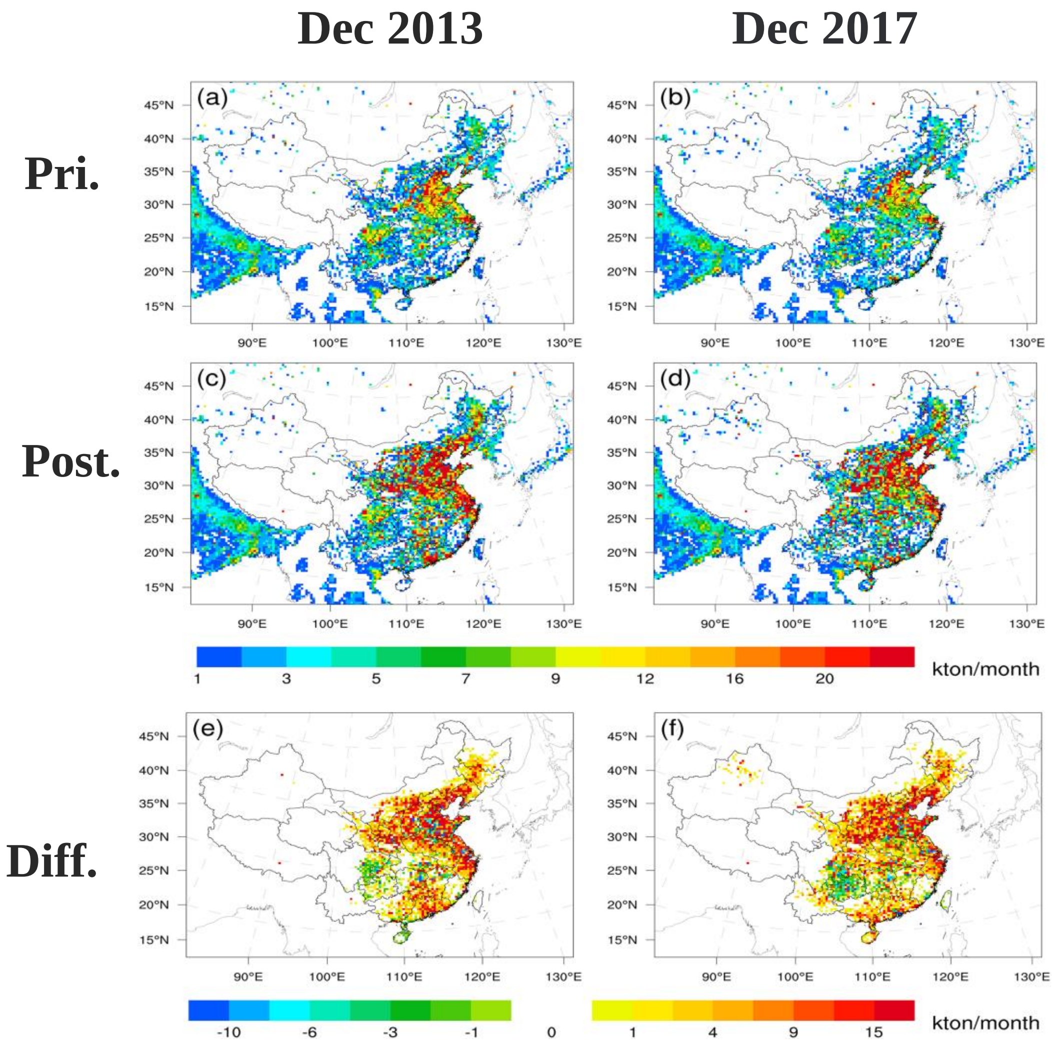


The frequency distribution of the relative biases

- ✓ Study period: December 2016
- ✓ The forecasting results over the entire 72 hours are improved, with mean RMSE decreased by 45%, 57%, 44%, 35% and 41% for CO, SO_2, NO_2, $PM_{2.5}$ and PM_{10}.
- ✓ The relative biases are more concentrated, mean biases decreased from -53%, 47%, -7%, -26% and -46% to -4%, -2%, -12%, -7% and -4%.

3. Applications

CO emission changes during the first stage clean air action (2013-2017)



- The CO emissions in December 2013 and 2017 are optimized using the observations in the corresponding period. The same sites were adopted.
- The spatial patterns of the increases in the two periods are similar
- For Mainland China, NCP, YRD, and PRD
  - ✓ Dec 2013: increased by 186%, 174%, 168% and 210%
  - ✓ Dec 2017: increased by 178%, 183%, 126% and 109%
- With posterior emissions, the performance of the simulated CO in Decembers of 2013 and 2017 are comparable.

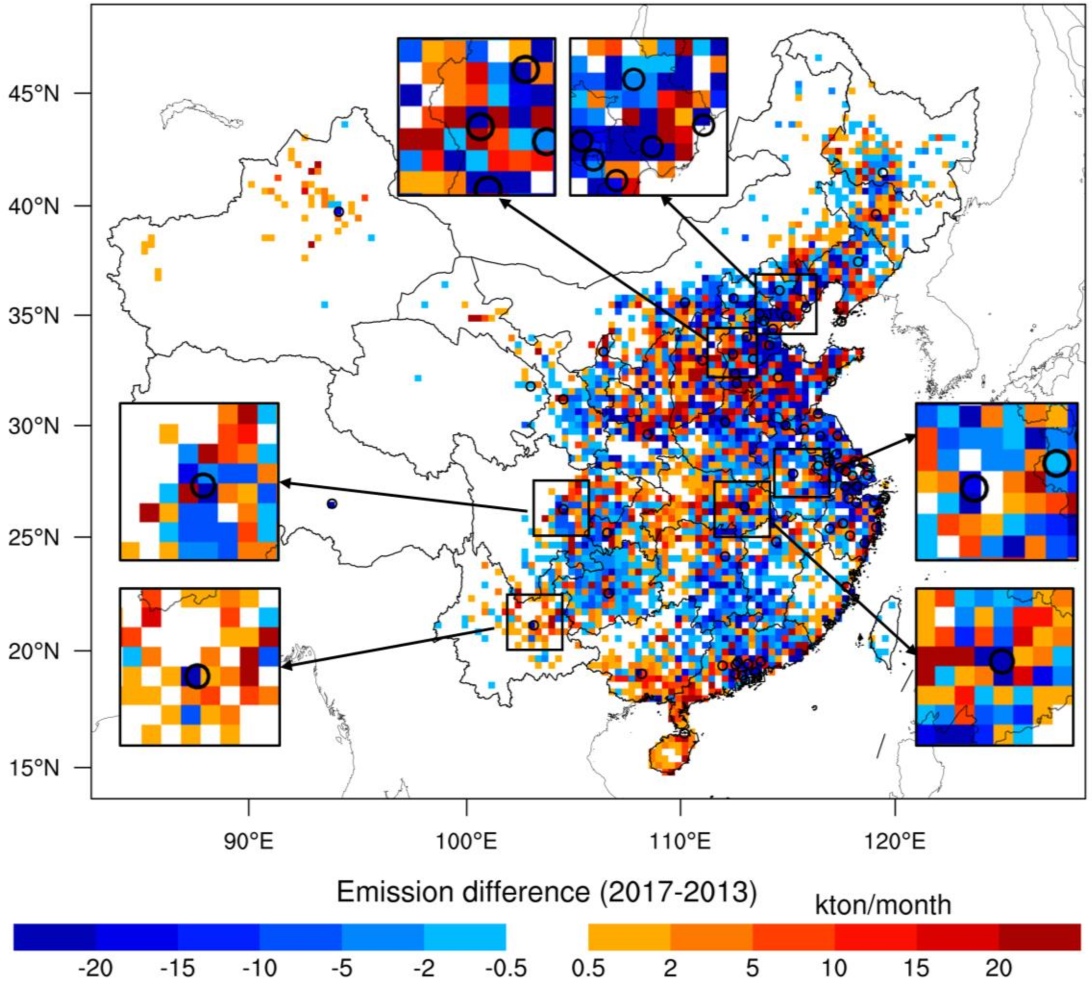
Table 5
Statistics Comparing the CO Concentrations From the Simulations With Prior and Posterior Emissions Against the Independent Observations in January 2014 and 2018

Region	Mean Obs. (mg/m^3)	Mean Sim. (mg/m^3)		BIAS (mg/m^3)		RMSE (mg/m^3)		CORR	
		CEPJ	VEPJ	CEPJ	VEPJ	CEPJ	VEPJ	CEPJ	VEPJ
January 2014									
Mainland	1.64	0.71	1.59	−0.94	−0.05	1.31	0.88	0.37	0.55
NCP	2.30	0.87	2.05	−1.43	−0.25	1.79	1.02	0.46	0.56
YRD	1.15	0.75	1.59	−0.40	0.44	0.62	0.83	0.49	0.49
PRD	1.29	0.54	1.25	−0.75	−0.04	0.88	0.53	0.24	0.28
January 2018									
Mainland	1.29	0.56	1.31	−0.73	0.02	0.90	0.55	0.36	0.61
NCP	1.54	0.63	1.45	−0.91	−0.09	1.11	0.62	0.45	0.62
YRD	1.12	0.67	1.31	−0.45	0.20	0.54	0.46	0.63	0.60
PRD	1.04	0.50	1.06	−0.54	0.03	0.60	0.28	0.30	0.59

3. Applications

CO emission changes during the first stage clean air action

Emissions changes between December 2013 and 2017



Emission changes based on inverted emissions

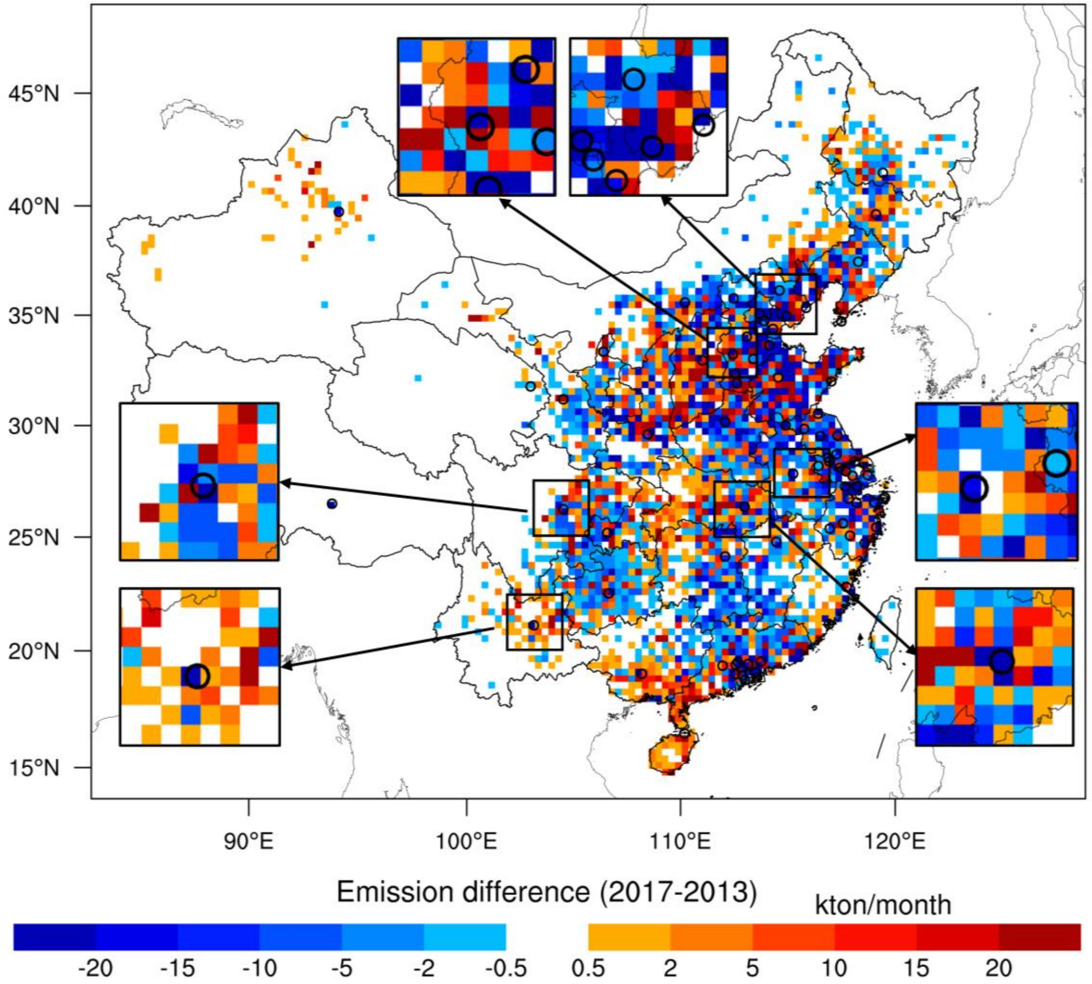
- The posterior CO emissions in 2017 are 17% lower than those in 2013
- Emission decreases in most key urban areas and developed regions, while increases in surrounding areas and certain central and western regions

Suggesting a emission migration from developed regions or urban areas to developing regions or surrounding areas.

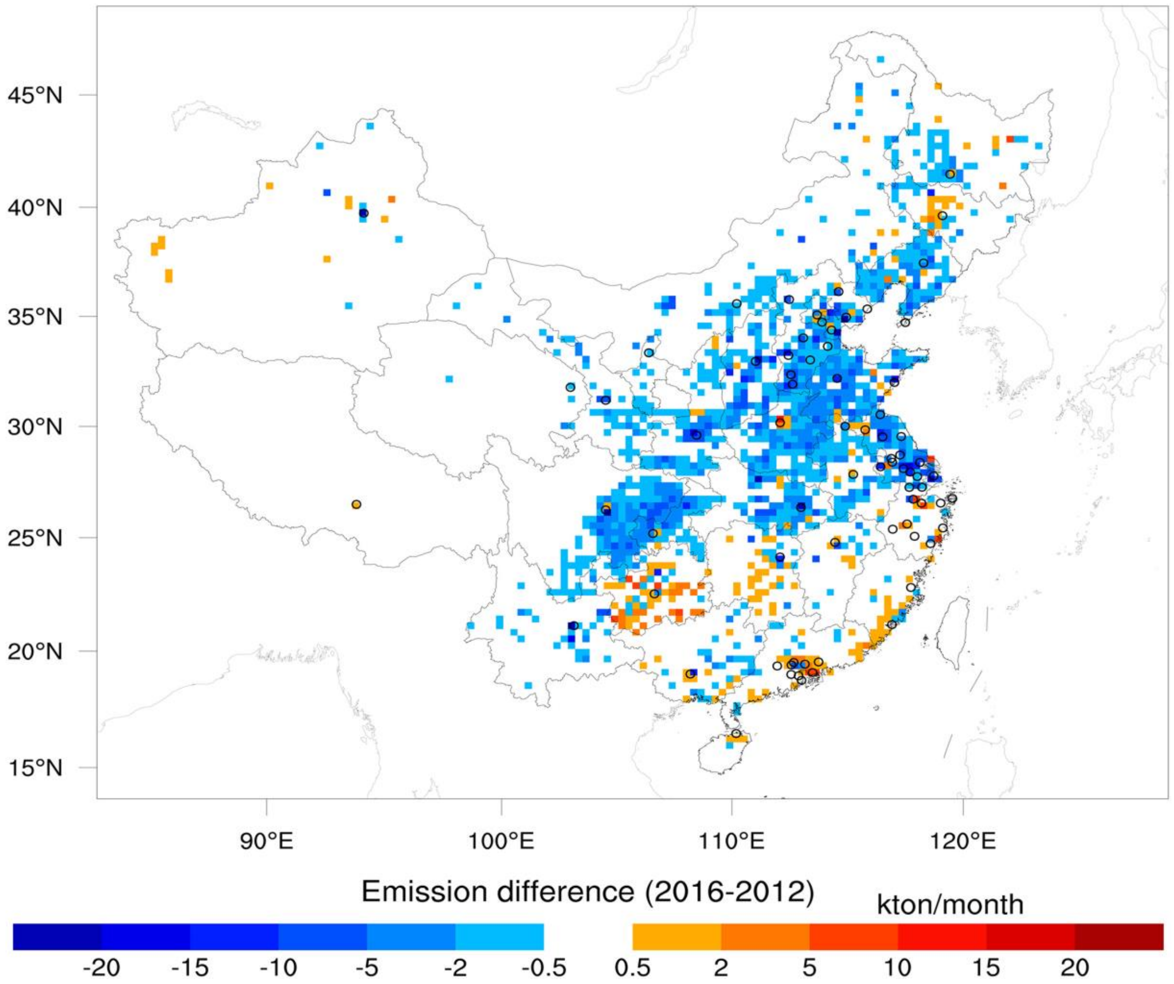
3. Applications

CO emission changes during the first stage clean air action

Emissions changes between December 2013 and 2017



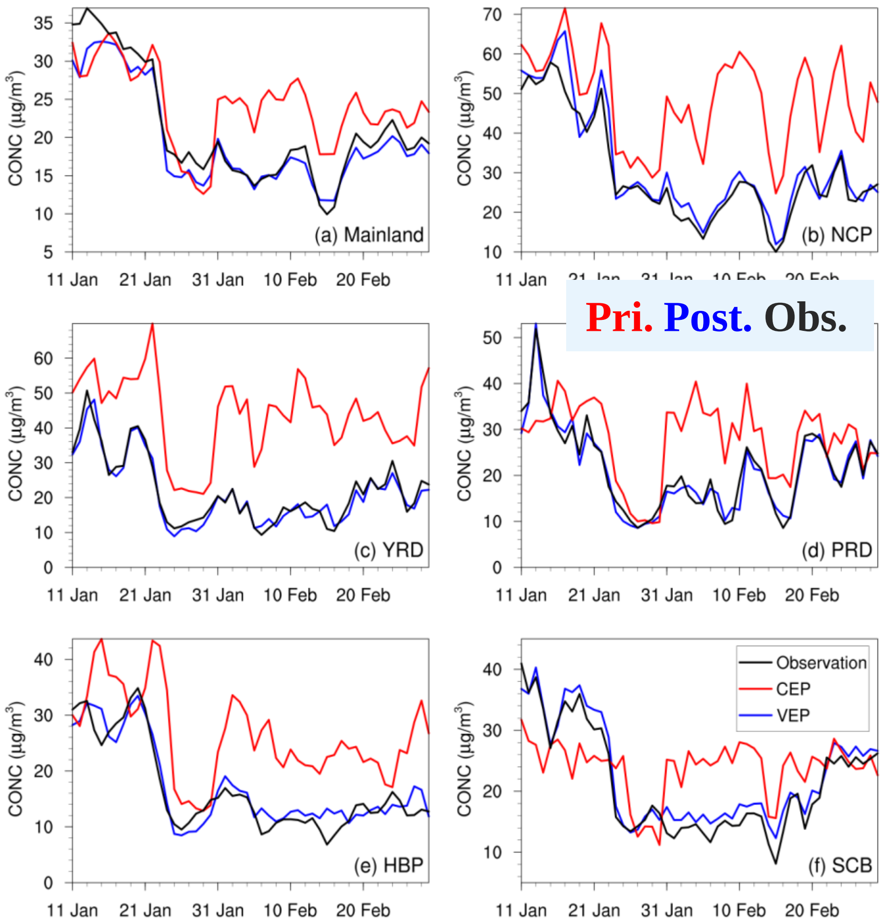
Emission changes based on inverted emissions



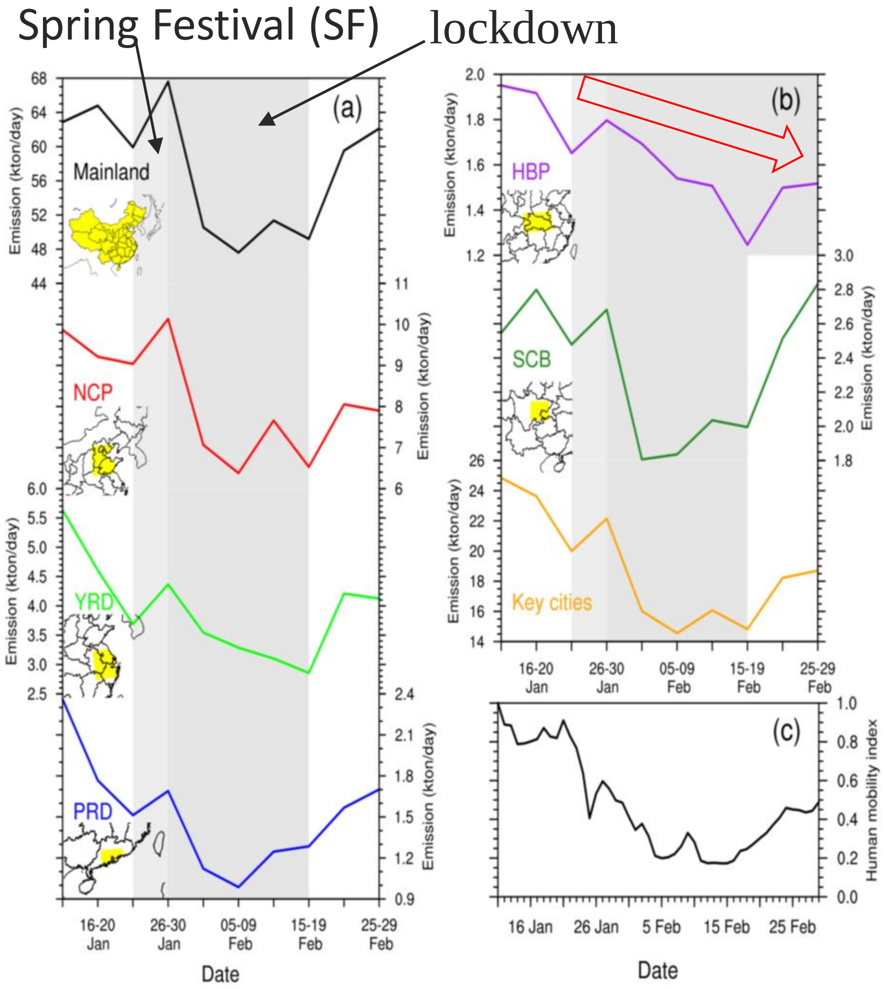
sion changes based on prior emission inventories

3. Applications

NOx emission changes during the COVID-19 lockdown in 2020



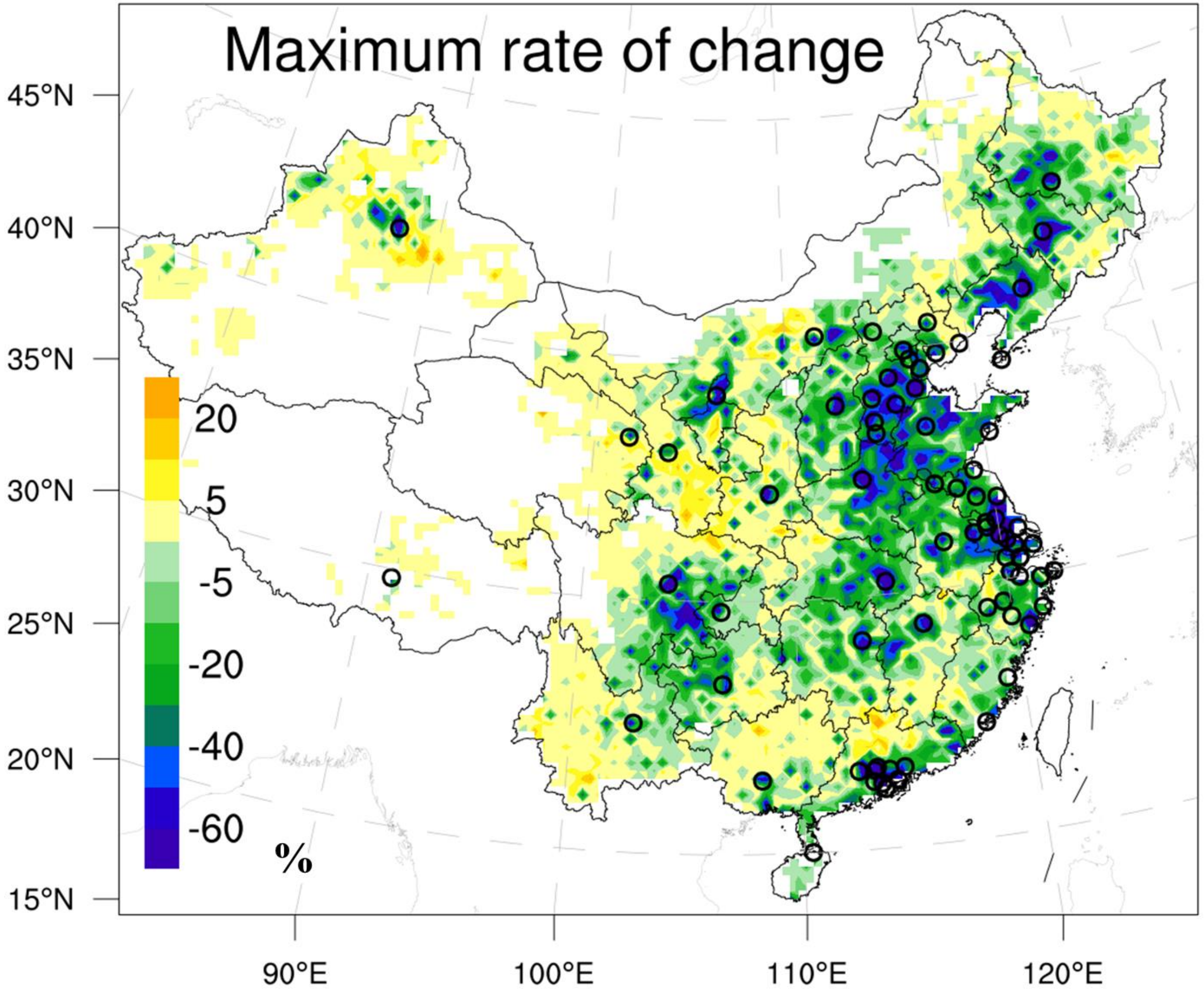
Daily simulated and observed NO_2 concentrations



Time series of 5-day averaged posterior emissions

- Daily NOx emissions are inferred using RAPAS and hourly NO_2 observations over China from 10 Jan to 28 Feb, 2020.
- The simulated NO2 with posterior emissions well match the observations.
- Two obvious processes of an initial decrease and subsequent increase in NOx emissions.
  - ✓ The first one, spring festival, NOx decrease is because of holiday, increase is due to fireworks.
  - ✓ The second one, lockdown

3. Applications NOx emission changes during the COVID-19 lockdown in 2020



Maximum emission reduction (%)

Compared with the emissions during 11-20 Jan, 2020

- Fell by more than 60% in many large cities and ~30% most small to medium cities
- Fell by 47% in the 74 key cities
- Slightly increased in certain remote areas, probably due to the return of workers from urban to rural, which increase the residential emissions in those areas.

4. Summary and limitations

1. We constructed a Regional multi-Air Pollutant Assimilation System (RAPASv1.0) based on the WRF/CMAQ model, 3DVAR and EnKF algorithm, which can optimize gridded and daily emissions of CO, SO_2, NO_x, primary $PM_{2.5}$ and coarse PM10 on regional scale by simultaneously assimilating hourly in-situ measurements.

2. Limitations

   1. **A idea profile is used for chemical boundary conditions, and no observation outside China is used**, which may leads to an overestimated emissions in China, especially for long lived species.

   2. **Model error is not considered**, all model-mismatch error is attributed to the uncertainty in emissions.

   3. The WRF-CMAQ model (off-line version) used in this system does **not consider the feedbacks from chemistry to meteorology**, which may also results in an overestimation of emissions, especially during winter.

   4. Primary $PM_{2.5}$ may be still overestimated due to **no observations of NH_3 and VOCs**, more evaluations with $PM_{2.5}$ components are needed.

   5. **Wind-blown sand and dust was not calculated**, resulting in significantly overestimated coarse PM_{10} emissions.

Thanks for your attention

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