



Supplement of

Changes in air pollutant emissions in China during two clean-air action periods derived from the newly developed Inversed Emission Inventory for Chinese Air Quality (CAQIEI)

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Supplementary Material

3 Text S1: Evaluation of the meteorological simulation

4 The performance of meteorological simulation is important for the inversion estimation since the meteorological parameters 5 influence the transport, chemical and removal process of air pollutants and affect the estimation of flow-dependent background error 6 covariance. Figure S18-S23 presents the comparisons of the simulated meteorological parameters, including zonal wind (U), 7 meridional wind (V), temperature (T), relative humidity (RH) and precipitation, against the observations obtained from China 8 Meteorological Administration (Figure S24). Evaluation statistics of meteorological simulation are also presented in Table S1. It 9 shows that the WRF simulation can generally captured the main features of the different meteorological parameters over the different 10 regions of China. The calculated correlation coefficient is 0.49-1.00 for different parameters, and the values of MB (RMSE) are -0.36-0.01 (0.3-0.52) m/s for U, -0.37-0.32 (0.32-0.80) m/s for V, -1.11-1.11 (0.6-2.17) °C for T, -11.2 to -2.59 (6.94-12.06) % 11 12 for RH, and -2.05-37.35 (5.45-61.62) mm for precipitation. This suggests WRF simulation generally well reproduce the 13 meteorological conditions for all regions of China, which is acceptable for the inversion estimates.

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15 Text S2: Assessment of the influences of site differences on the emission inversions

16 The emission increments at the observation sites (a posteriori minus a priori) for different species in China from 2013 to 2015 17 under the scenarios of fixed observation sites (blue lines) and varying observation sites (orange) were calculated to assess the 18 influences of the site differences on the emission inversions (Fig. S2). In the fixed-site scenario, it is assumed that the number of 19 observation sites remains constant at the 2013 level while in the varying-site scenario, the number of observation sites increases 20 over time. The differences in emission increments between these two scenarios are used to analyze the impact of changes in the 21 observation coverage on the emission inversions. Please note that, to simplify calculations, we only computed the emission 22 increments at the locations of observation sites. Therefore, they may not be equal to the emission increments calculated for the entire 23 grid as reported in the paper. However, they are still useful indicators for the effects of emission inversion. In addition, since we did 24 not consider the temporal variation in the a priori emissions, the changes of the emission increments can be used to approximate the 25 temporal variations of the a posterior emissions. It can be clearly seen that that there are obvious differences in the emission 26 increments between the two scenarios. The emission increment is larger in the varying-site scenario than that in the fixed-site 27 scenario for all species due to the increases of observation sites. Moreover, as indicated in Fig. S2, the changes of observation sites 28 were shown to significantly affect the estimation of the emission trend in 2013 and 2014. Most of species showed decreasing trends in their inversed emission under the fixed-site scenario. However, under the varying-site scenario, the decreasing trends were smaller 29 for PM_{2.5}, NO_x and NMVOC, and the emissions of PM₁₀ and CO even showed increasing trends. This is due to that the emission 30 31 increments were positive over most of observation sites for these species as demonstrated in Fig.3. Thus, the increases of observation 32 site would lead to increases of positive emission increments and higher a posteriori emissions, which may counteract the decreasing 33 trends or even lead to an opposite trend. These results provide the evidences that the increasing trends in the total emissions of PM_{10} 34 and CO from 2013 to 2015 seen in Fig. 6 and Fig. 7 are highly likely to be a spurious trend caused by the changes of observation 35 coverage. The weak emission changes in $PM_{2.5}$ and NO_x (Fig. 6 and Fig. 7) may also be related to the changes in the number of 36 observation sites. The SO₂ emission is an except that its calculated trend is larger under the varying-site scenario than that under the 37 fixed-site scenario. This is because that the emission increment for the SO_2 is generally negative over the most sites, thus the 38 increased observation sites would lead to larger decreasing trend in the inversed emissions of SO₂. To date, these results highlighted 39 the significant influences of the site differences on the estimated emissions and their trends. Therefore, we recommend not to use 40 the emission in 2013 and 2014 when analyze the trends of the emissions.

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Text S3: Comparisons of model performance driven by CAQIEI with that driven by more recent bottom-up emission inventories

44 To obtain a better understanding of the accuracy of our inverse emission inventory, we conducted a one-year simulation of air pollution in China for year 2020 with more recent bottom-up emission inventories and compared its performance with that driven 45 by the CAQIEI. The used bottom-up inventories in this simulation case includes the HTAPv3 (Crippa et al., 2023) inventory for the 46 47 anthropogenic emissions outside China with a base year of 2018; the MEIC inventory for the anthropogenic emissions over China with a base year of 2020; the CAMS emission inventory (https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-48 49 emission-inventories?tab=overview, last access: 19 June 2024) for the biogenic, soil and oceanic emissions; and the Global Fire 50 Assimilation System (GFAS) (Kaiser et al., 2012) for the biomass burning emissions. Note that since the MEIC emission inventory does not include the ship, air and waste emissions. Emissions from these sectors over China were provided by the HTAPv3 emission 51 52 inventory. For clarity, in following content, we name this simulation case as the MEIC-HTAPv3 based on the anthropogenic 53 emission inventory used.

54 Figure S25 shows the time series of hourly concentrations of different air pollutants in China obtained from observation and 55 simulation driven by the CAQIEI and more recent bottom-up inventories. Comparisons of the evaluation statistics of these two 56 simulation scenarios are also presented in Table S3. It shows that updating the bottom-up emission inventories to a more recent year 57 does improve the model performance compared to the outdated a priori emission inventory (Table 2), suggesting that the bottom-58 up emission inventory has to some extent captured the changes of air pollutant emissions in China. It is also encouraging to find 59 that the model performance driven by CAQIEI and MEIC-HTAPv3 is similar for the PM_{2.5}, PM₁₀, and SO₂ over the NCP, NE, SE 60 and SW regions, both significantly improved from the a priori emission inventory. This suggest that both the top-down and recent bottom-up emission inventories have good performance in capturing the emission changes of these species over these regions and 61 62 they yield consistent estimations. However, the model simulation driven by MEIC-HTAPv3 still have negative biases in the CO 63 concentrations possibly due to the underestimations of CO emissions as we illustrated in Sect.4.3.1.3. Similarly, due to the errors in

64	the dust emission, there are negative biases in the simulated $PM_{2.5}$ and PM_{10} concentrations over the western China driven by MEIC-
65	HTAPv3. On the contrary, the simulated NO ₂ concentrations in MEIC-HTAPv3 are higher than the observations over the NCP, NE
66	and SE regions, which also partly contributes to the underestimated O3 concentrations over these regions. The CAQIEI generally
67	achieves better performance in simulating the air pollutant concentrations in China as indicated by higher values of correlation
68	coefficient and lower values of bias and root mean square of error in the model simulation driven by CAQIEI than that driven by
69	MEIC-HTAPv3 (Table S3).
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97 Table S1: Evaluation statistics for the meteorology simulation

Region		U (m/s)		V (m/s)		T (°C)			RH (%))		Precipitat mm/moi	
	R	MB	RMSE	R	MB	RMSE									
NCP	0.95	0.01	0.30	0.95	-0.02	0.49	1.00	-0.42	0.84	0.95	-11.24	11.66	0.95	3.74	18.56
NE	0.94	0.37	0.51	0.89	-0.08	0.49	0.99	-1.11	2.17	0.77	-2.59	7.18	0.97	12.09	19.76
SE	0.84	-0.27	0.37	0.98	-0.37	0.80	1.00	-0.40	0.60	0.88	-7.00	7.58	0.94	37.35	61.62
SW	0.63	-0.44	0.52	0.69	0.04	0.37	0.99	1.11	1.27	0.87	-5.84	6.94	0.92	16.85	40.18
NW	0.49	-0.36	0.51	0.58	0.32	0.43	0.99	0.83	1.91	0.79	-9.49	12.06	0.51	-2.05	5.45
CENTRAL	0.95	0.10	0.41	0.70	-0.08	0.32	1.00	-0.27	0.93	0.85	-8.59	10.30	0.97	4.64	10.87

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99 Table S2 The average mean (standard deviation) of the calculated factor for the inflation of the ensemble member over

NCP NE SE SV	W NW Central
PM _{2.5} 1.0 (0.2) 1.7 (1.6) 1.0 (0.0) 6.8 ((8.5) 3.1 (3.8) 3.9 (3.9)
PM_{10} 1.4 (0.7)7.2 (8.0)2.4 (0.8)78.1 (1.1)	108.2) 26.3 (36.5) 36.0 (49.0)
SO ₂ 1.4 (0.7) 4.1 (3.2) 2.3 (0.8) 176.1 ((254.6) 7.8 (6.5) 58.6 (72.5)
NO _x 1.0 (0.1) 1.7 (0.7) 1.2 (0.3) 8.1 ((5.3) 2.8 (1.3) 5.4 (4.1)
CO 1.0 (0.1) 2.8 (2.3) 1.4 (0.4) 18.8 ((16.8) 6.8 (6.9) 8.6 (10.0)
NMVOC 1.4 (0.6) 4.5 (4.4) 1.6 (0.5) 8.1 ((8.6) 6.5 (5.8) 8.1 (10.1)

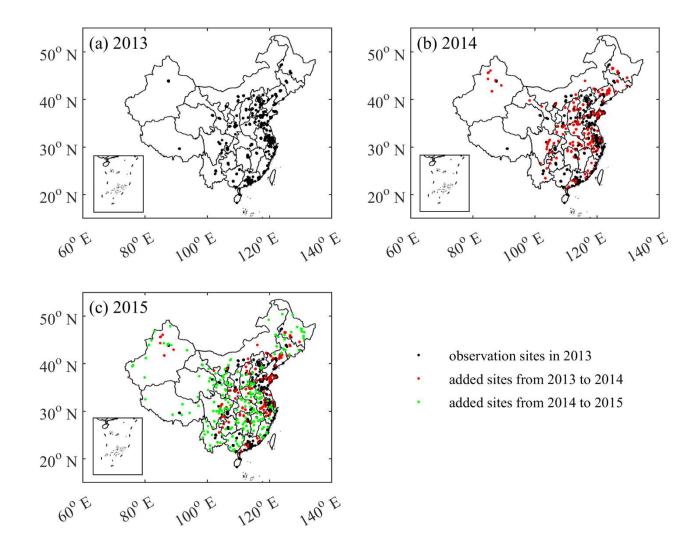
100 different regions of China for different species

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Table S3: Evaluation statistics of the model simulation driven by CAQIEI (outside brackets) and more recent bottom-up
 inventories (inside brackets) in 2020

PM _{2.5}	PM_{10}	SO_2	NO ₂	CO	O_3
$(\mu g/m^3)$	$(\mu g/m^3)$	$(\mu g/m^3)$	$(\mu g/m^3)$	(mg/m^3)	$(\mu g/m^3)$
0.77 (0.53)	0.73 (0.44)	0.37 (0.19)	0.69 (0.45)	0.67 (0.40)	0.75 (0.48)
3.6 (5.3)	-0.3 (-14.9)	0.3 (0.7)	-0.9 (6.7)	-0.06 (-0.4)	6.3 (-13.7)
10.5 (15.8)	-0.5 (-25.9)	2.6 (7.6)	-3.4 (26.2)	-8.9 (-52.7)	10.2 (-22.1)
24.6 (34.2)	37.4 (49.1)	10.9 (13.6)	15.9 (25.1)	0.4 (0.6)	30.3 (42.3)
	$(\mu g/m^3)$ 0.77 (0.53) 3.6 (5.3) 10.5 (15.8)	$(\mu g/m^3)$ $(\mu g/m^3)$ 0.77 (0.53)0.73 (0.44)3.6 (5.3)-0.3 (-14.9)10.5 (15.8)-0.5 (-25.9)	$(\mu g/m^3)$ $(\mu g/m^3)$ $(\mu g/m^3)$ 0.77 (0.53)0.73 (0.44)0.37 (0.19)3.6 (5.3)-0.3 (-14.9)0.3 (0.7)10.5 (15.8)-0.5 (-25.9)2.6 (7.6)	$(\mu g/m^3)$ $(\mu g/m^3)$ $(\mu g/m^3)$ $(\mu g/m^3)$ 0.77 (0.53)0.73 (0.44)0.37 (0.19)0.69 (0.45)3.6 (5.3)-0.3 (-14.9)0.3 (0.7)-0.9 (6.7)10.5 (15.8)-0.5 (-25.9)2.6 (7.6)-3.4 (26.2)	$(\mu g/m^3)$ $(\mu g/m^3)$ $(\mu g/m^3)$ $(\mu g/m^3)$ $(m g/m^3)$ 0.77 (0.53)0.73 (0.44)0.37 (0.19)0.69 (0.45)0.67 (0.40)3.6 (5.3)-0.3 (-14.9)0.3 (0.7)-0.9 (6.7)-0.06 (-0.4)10.5 (15.8)-0.5 (-25.9)2.6 (7.6)-3.4 (26.2)-8.9 (-52.7)



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Figure S1: Spatial distributions of observation sites in (a) 2013, (b) 2014 and (c) 2015. The observation sites in 2013 were marked as black dots, while the added observation sites from 2013 to 2014 and those from 2014 to 2015 were marked as red and green dots, respectively.

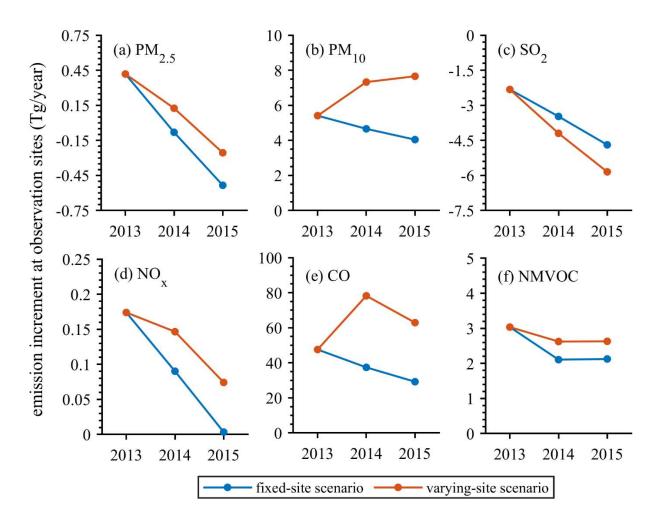
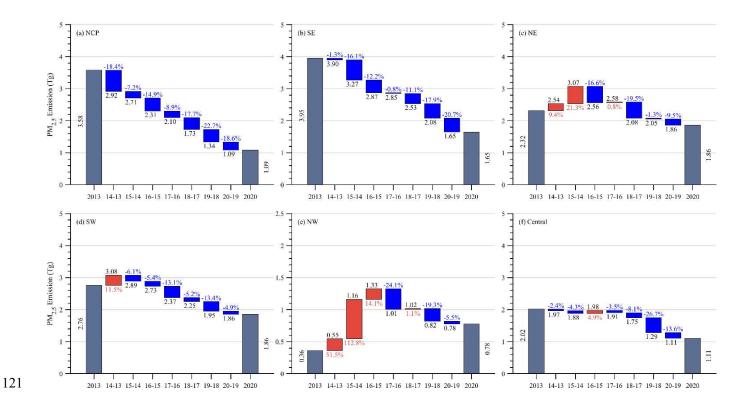
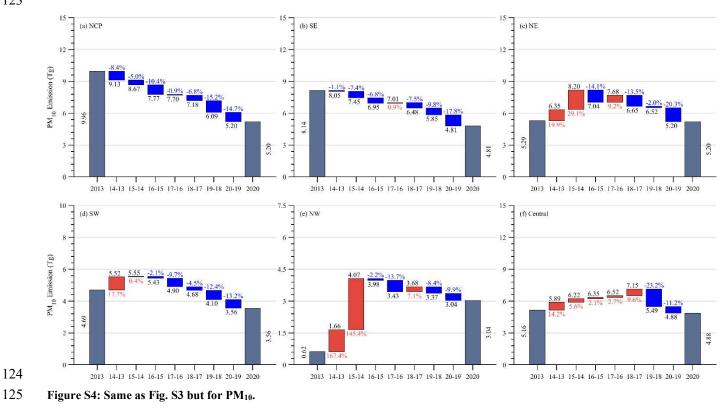
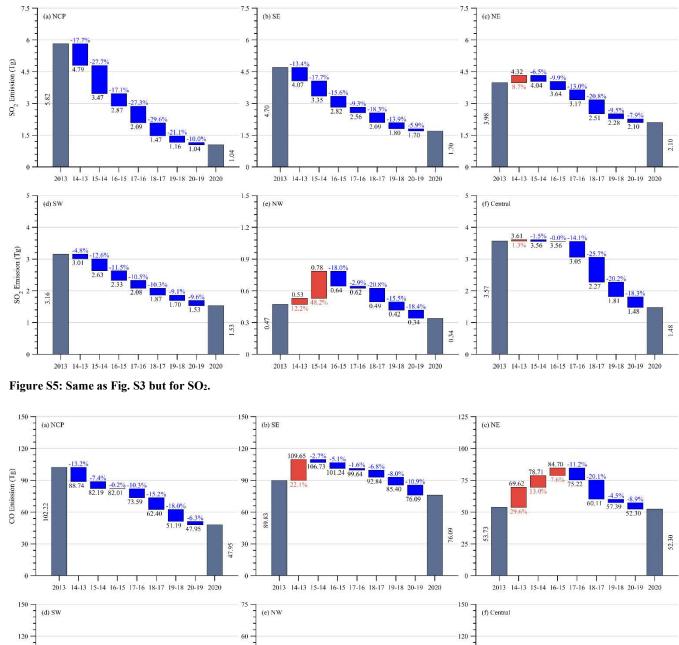


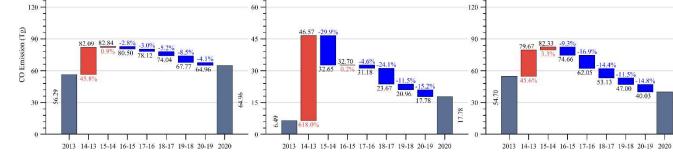
Figure S2: The calculated total emission increments at the observation sites for different species under the fixed-site scenario and varyingsite scenario.



122 Figure S3: Emission changes of PM_{2.5} from 2013 to 2020 over different regions of China obtained from CAQIEI.



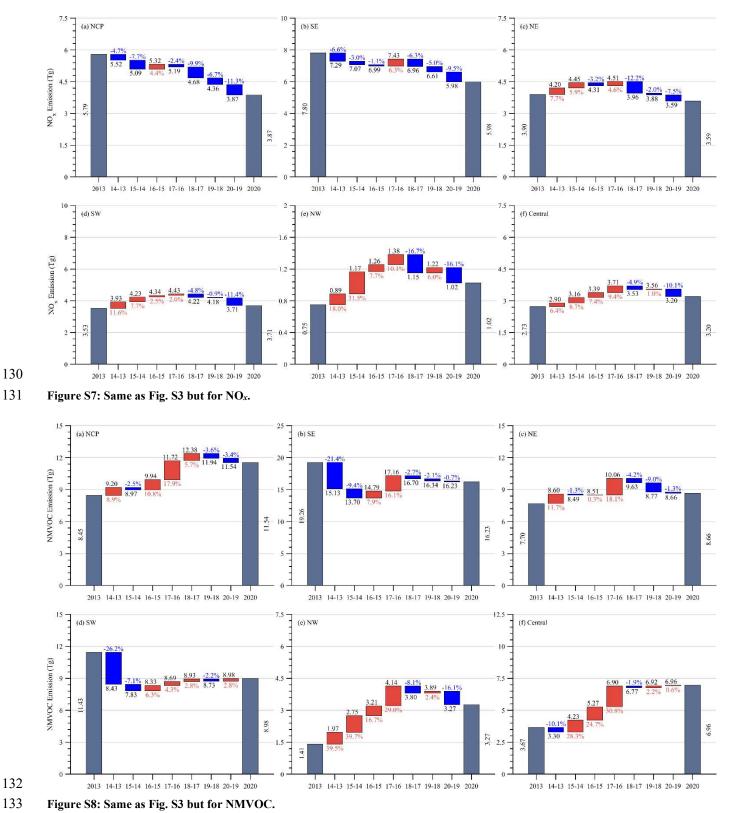


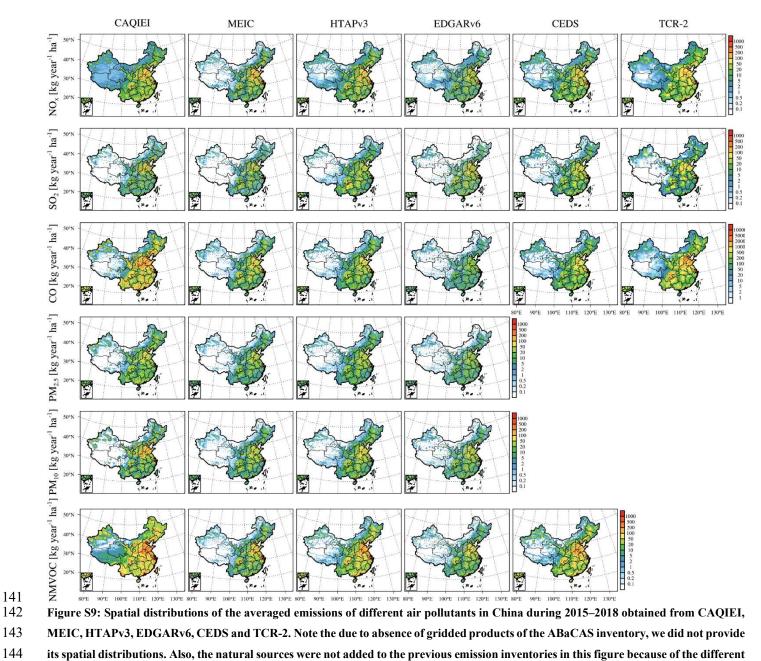


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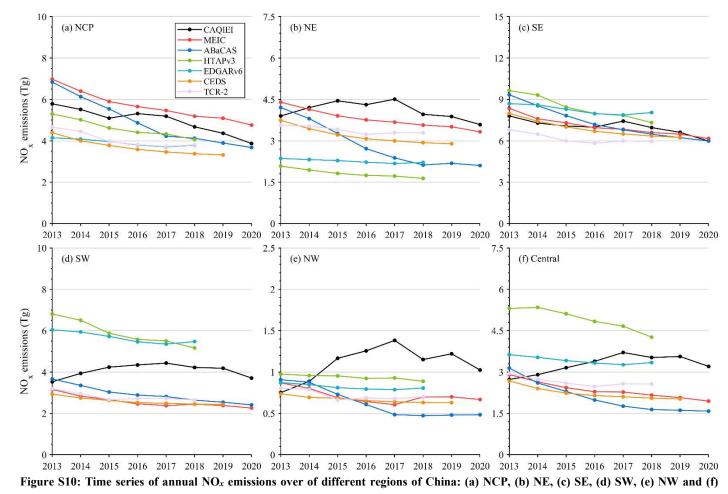
129 Figure S6: Same as Fig. S3 but for CO.

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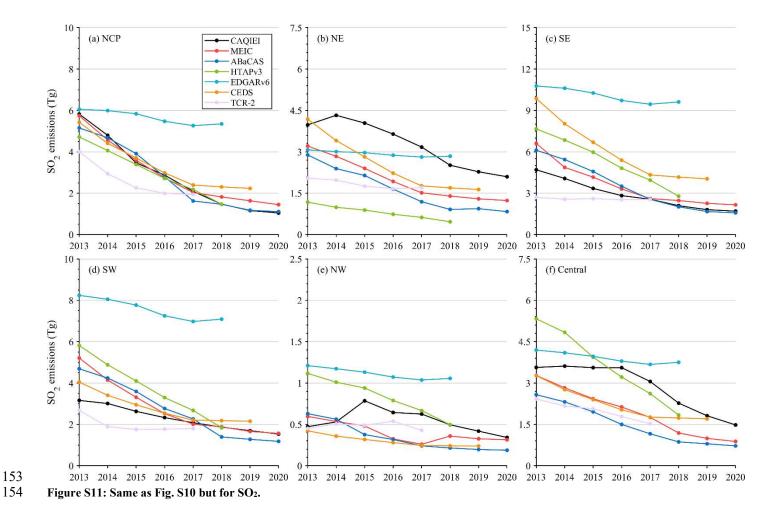


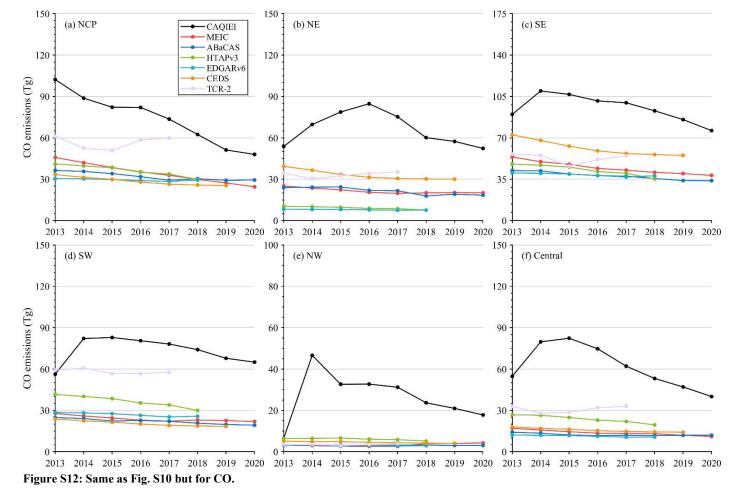


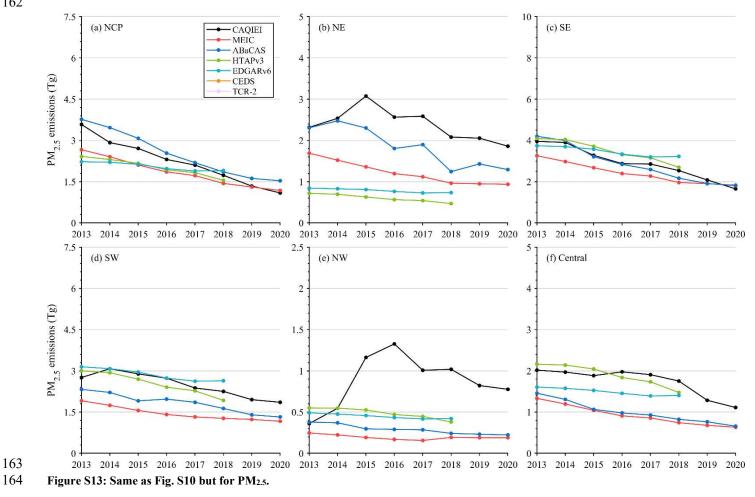
- spatial resolutions among these inventories.

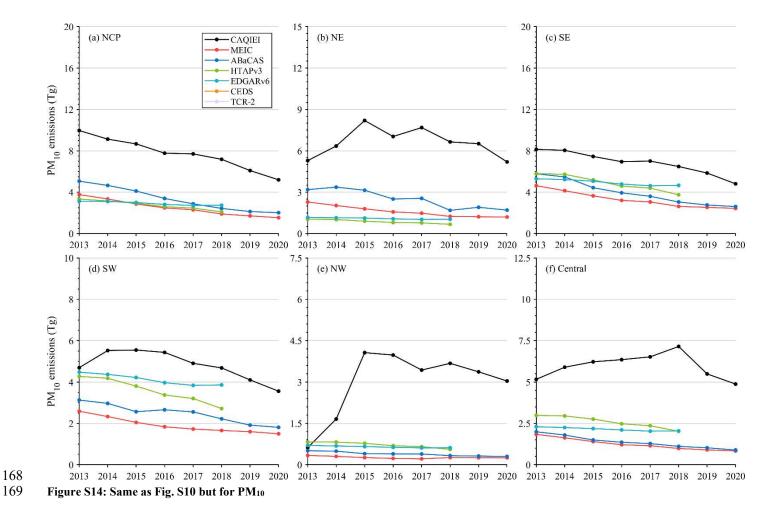


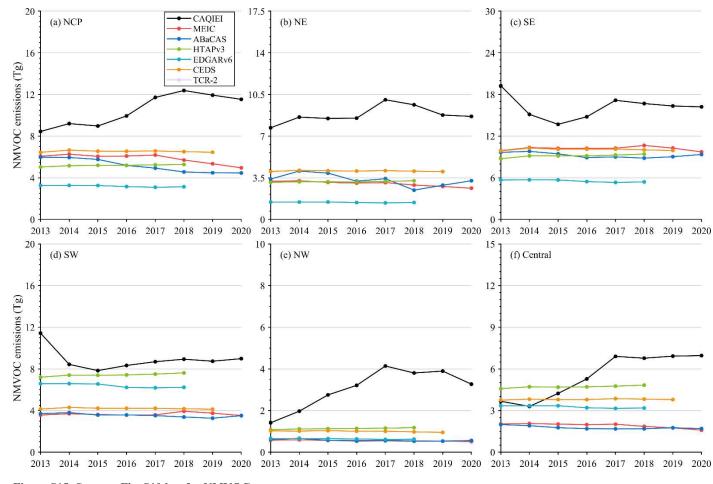
149 Central from 2013 to 2020 obtained from CAQIEI and previous inventories. Note that the natural sources were not included in the 150 previous inventories in this figure.











173 Figure S15: Same as Fig. S10 but for NMVOC.

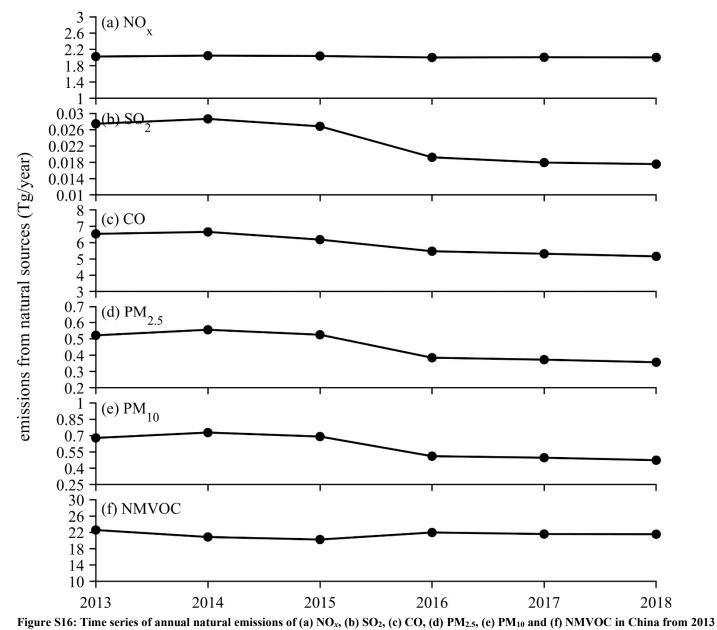
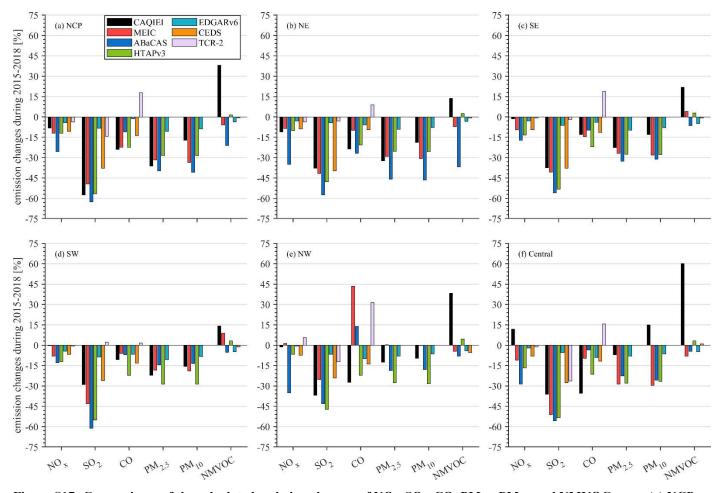


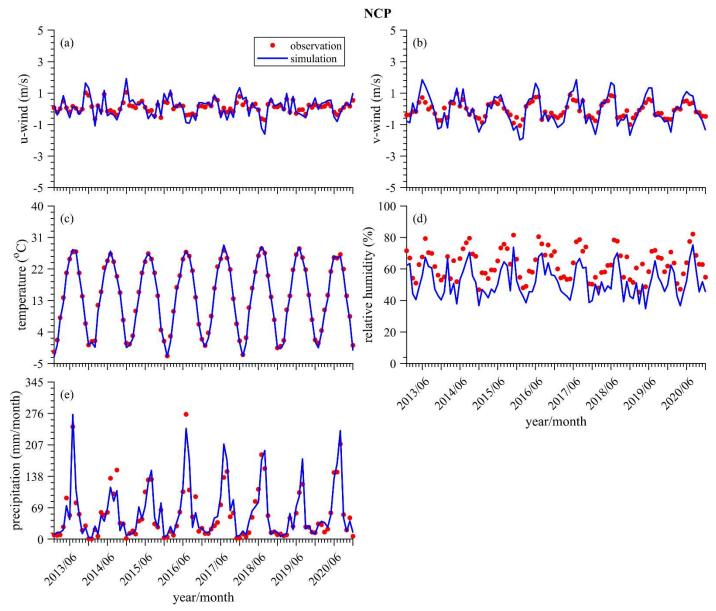
Figure S16: Time series of annual natural emissions of (a) NO_x , (b) SO_2 , (c) CO, (d) $PM_{2.5}$, (e) PM_{10} and (f) NMVOC in China from to 2018. The considered natural sources includes the biogenic, biomass burning and soil emissions.



180 Figure S17: Comparisons of the calculated emission changes of NO_x, SO₂, CO, PM_{2.5}, PM₁₀, and NMVOCs over (a) NCP,

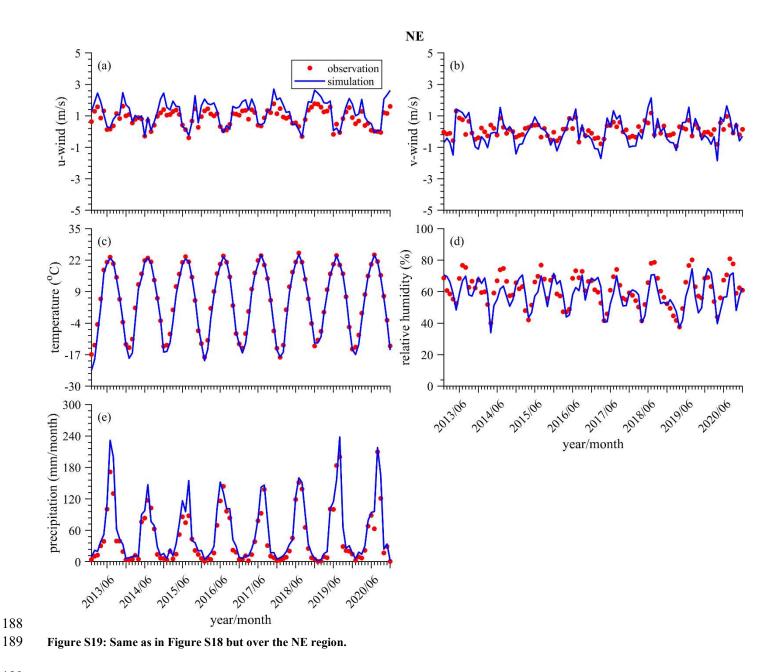
(b) NE, (c) SE, (d) SW, (e) NW and (f) Central regions of China from 2015 to 2018 between CAQIEI and previous
inventories.

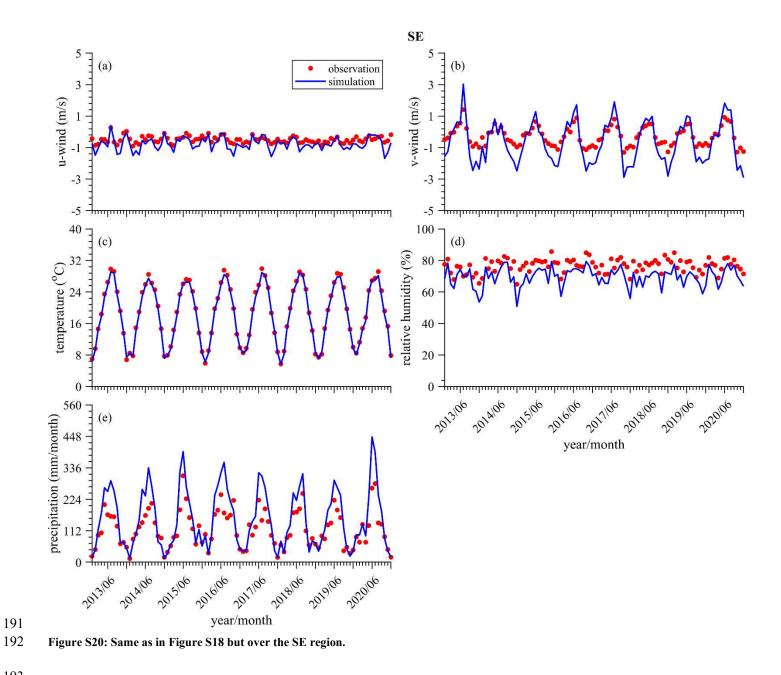
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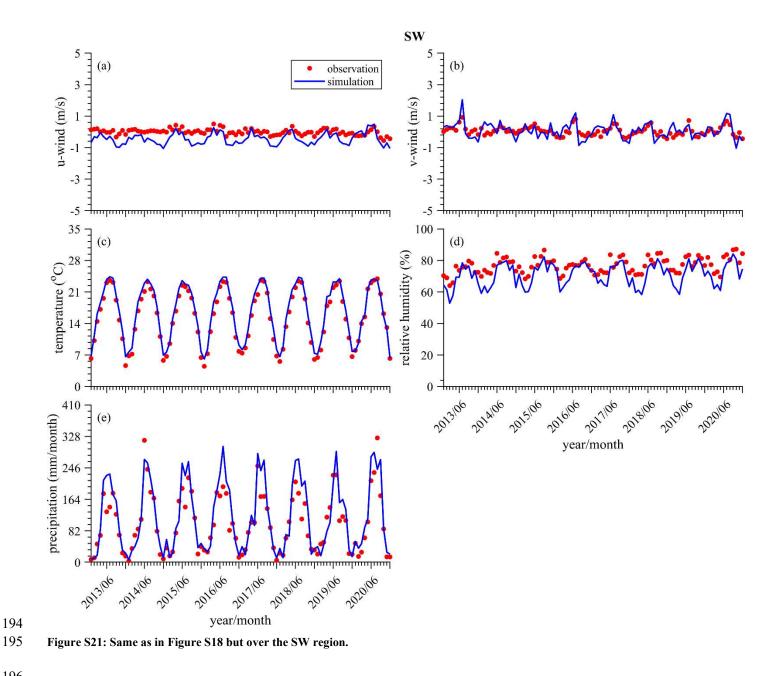


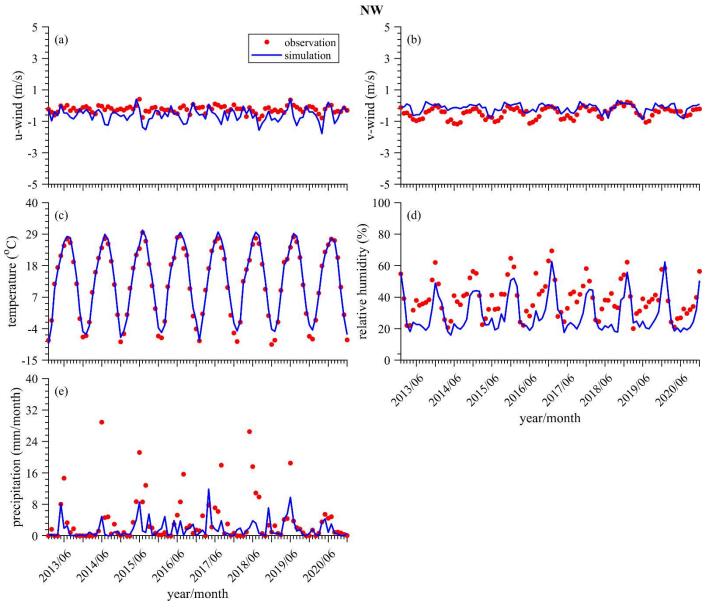
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Figure S18: Timeseries of observed (red dots) and simulated (blue line) monthly values of (a) zonal wind, (b) meridional wind, (c) temperature, (d) relative humidity and (e) precipitation over NCP region from Jan 2013 to Dec 2020.

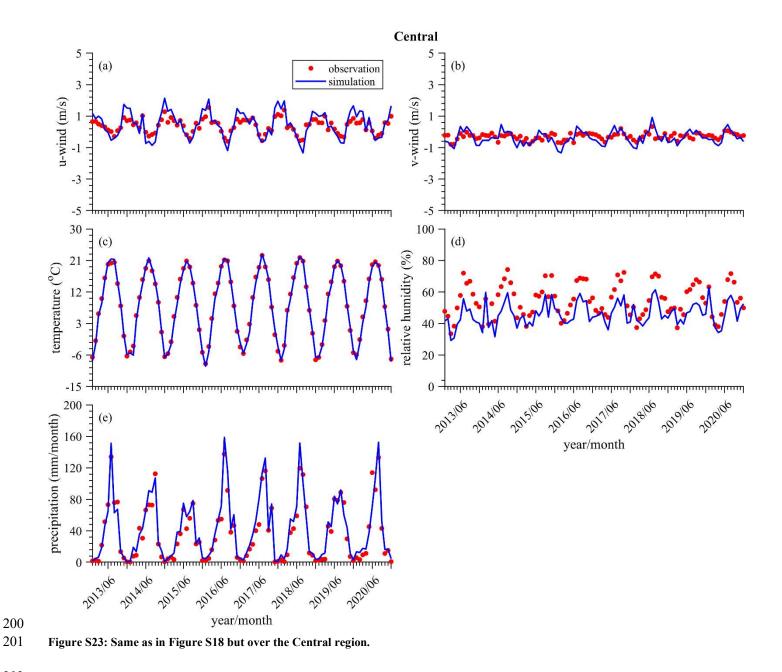


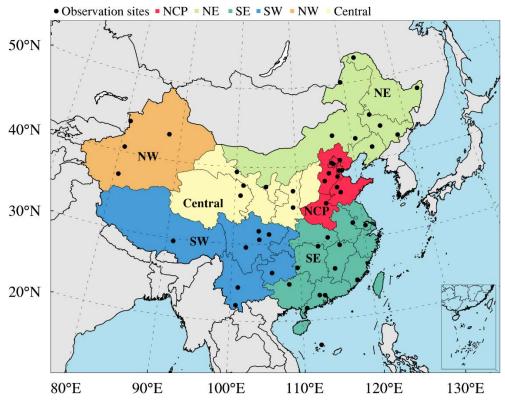






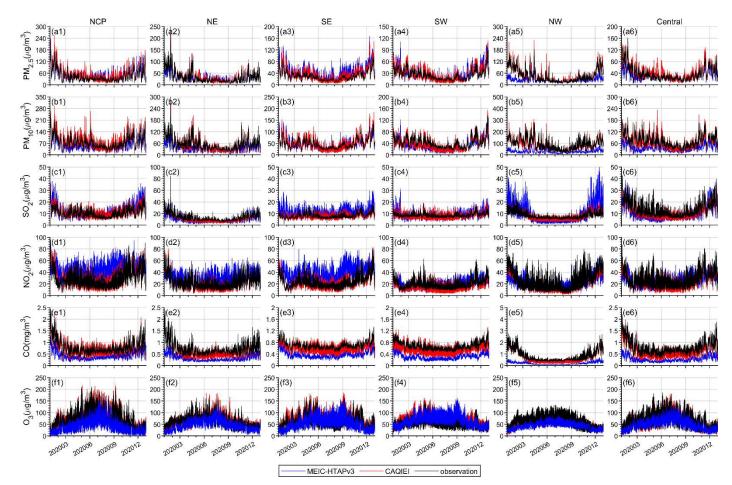
198 Figure S22: Same as in Figure S18 but over the NW region.





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Figure S24: Spatial distribution of meteorological observation sites used in the evaluation of meteorology simulations over different regions of China



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Figure S25: Timeseries of observed (black lines) and simulated concentrations of different air pollutants in China driven by CAQIEI (red
 lines) and MEIC-HTAPv3 (blue lines) over different regions of China.