***Supplementary Information*:**

Contrasting emission trends between the two phases of China's clean air actions from 2013–2020

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Table of Contents

Supplementary Method:

Updates in MEIC v1.4 based on the previous version

In MEIC v1.4, the major updates compared to the previous version (i.e., v1.3) were the inclusion of the unit-based cement emission model[1] and the survey-based residential emission model[2], as well as the updates of the available activity and emission control information from statistical yearbooks and the Ministry of Ecology and Environment (MEE, unpublished data, referred to hereafter as the MEE database).

In MEIC v1.4, a unit- and technology-based methodology was adopted for SO_2, NO_x, CO, $PM_{2.5}$, and PM_{10} emissions in the cement industry since the year 1990[1]. Cement production comprises three stages: raw material preparation, clinker calcination, and cement grinding. NO_x, SO_2 and CO are emitted from fuel combustion during the clinker calcination process, whose emissions were estimated using the coal consumed in the cement kilns. PM emissions involve the entire process of cement production, including both organized and fugitive emissions. PM emissions were calculated with a dynamic methodology to consider the evolution of PM control technologies in different cement kilns. Detailed unit-level data for 3125 clinker production lines and 4549 cement grinding stations were obtained from the MEE database, including clinker and cement production, production capacity, operating and retiring dates, PM and NO_x control technologies, and the locations of each unit. The unit-level coal consumption was calculated as the product of unit-based clinker production and annual energy intensity. We developed a linear regression model between the logarithm of energy intensity and time in years with all the available data taken from literature to predict the energy intensity in each year, which facilitate the calculation of coal use for each production line. Although the provincial activity data from statistics were used to constrain the unit-level activity rates, the estimated emissions using the unit-level model were different from our previous estimates on provincial scale[3], which could partially explain the differences in the industrial sectors between MEIC v1.3 and v1.4.

Besides, MEIC v1.4 adopted a survey-based solid fuel consumption model to estimate the emissions from rural residential[2]. Inadequate statistics for residential solid fuel consumption led to large uncertainties in emission estimates in the residential sectors. To address this gap, our previous study conducted a nationwide survey of fuel consumption rates, use patterns, and combustion technologies of solid fuels in rural areas. The field survey covered ~17,000 rural residential households in 183 randomly selected counties in China. Then Generalized Additive Models (GAMs) were developed to estimate per capita coal and biofuel consumption at the county level, respectively, which established the non-linear relationship between solid fuel use, heating degree days (HDD), income, coal production, coal price, and vegetation coverage. By using the GAMs, county-level coal and biofuel consumption for each year were estimates. The survey-based solid fuel consumption model estimated a ~60% higher coal consumption and a ~50% lower crop residue consumption in Chinese rural households, compared to those reported in official energy statistics[2]. This biases in official data was also discussed in other studies conducting field survey over China[4]. Then, the survey-based estimates of coal and biofuel use were utilized to calculate air pollutant emissions, which introduced large differences when comparing against MEIC v1.3 that used official energy statistics.

In addition to the improvements in the emission models, the input data (e.g., activity rates, emission factors and technology penetration rates) were also updated in the new version. First, detailed activity rates for each province in China after the year 2016 were not available when MEIC v1.3 emissions were developed[3]. Instead, province-level activity rates after 2016 were extrapolated using national activity data and weighting factors adopted from previous years. In MEIC v1.4, those extrapolated activity data were replaced by actual provincial data obtained from statistical yearbooks. Meanwhile, the penetrations of control technologies and relevant removal efficiencies for the power and industrial sectors were also updated for these years, as more unit-level information were available from the MEE database. Second, emission factors for NMVOC were systematically updated according to our most recent study[5], which affected NMVOC emission estimates for all years. Briefly, emission factors were compiled based on measurements or surveys, including the European Environment Agency (EEA) guidebook[6], the AP-42 database[7] and peer-reviewed literatures. More details about the values and sources of the NMVOC emission factors could be found in Li et al.[5]. Third, SO_2 emission factors for the on-road vehicles were updated compared to the previous version, as the new version has taken the impact of tightened fuel standards into consideration. However, such updates would not greatly change the SO_2 emission estimates as the contribution from on-road vehicles to SO_2 was minor.

Comparison with MEIC v1.3

Supplementary Figure 6 compares the anthropogenic emissions of major air pollutants from MEIC v1.3 and v1.4 during 2013–2017, when the two versions of emissions were overlapped in time. Overall, national total emissions agree well between different versions, although small discrepancies exist. The differences in NMVOC between the two versions were mainly caused by the updated emission factors for NMVOC in v1.4, as described above. The slight decrease in $PM_{2.5}$, PM_{10}, BC, and OC emissions in MEIC v1.4 compared to v1.3 were largely contributed by the residential sector, because the survey-based estimation of coal and biofuel consumption lead to a lower emission estimation. In addition, the updates of actual activity and emission control information for 2016 and 2017 in MEIC v1.4 also result in a small difference in SO_2 emissions over those years.

We also compared the estimated emission trend between the two version of emissions, as shown in Supplementary Table 2. The absolute and relative emission changes on national scale during 2013–2017 are comparable between versions. For example, SO_2 emission decreased by 14.9 Tg (59%) in MEIC v1.3, comparable to a 14.4 Tg (56%) decrease in MEIC v1.4. The discrepancies are larger at sector scale, especially for industry and residential sectors. This is because the major updates of emission models in MEIC v1.4, i.e., the unit-based cement model and the survey-based solid fuel model, happened in industrial and residential sectors.

Comparison with satellite and in situ observations

We adopt atmospheric observations to evaluate and validate the emission trends estimated in this study. We use NO_2 column retrievals from Level-3 Aura/OMI Global OMNO2d v003 Data Products and SO_2 column retrievals from Level-3 Aura/OMI Global OMISO2e v003 Data Products. The trends of satellite observations are calculated over eastern China (29°N–41°N, 108°E–123°E), where anthropogenic sources are dominant.

We also collect surface-level SO_2, NO_2 concentration data from national air quality monitoring stations (http://www.cnemc.cn/) for the period of 2013–2020. These real-time monitoring stations were established in 2013 and had the ability to report hourly concentrations of criteria pollutants. 496 sites in 74 cities in 2013 were established and then expanded to 1734 sites in 339 cities in 2020. The trend of SO_2 and NO_2 are averaged over 74 cities where the ground observations are continuous during 2013–2020.

Supplementary Figure 5 compares the estimated emission trends of SO_2 and NO_x with satellite retrievals and ground-based observations during 2013–2020. In general, trends in SO_2 and NO_x emissions are consistent with observations from both space and ground. We estimated that SO_2 and NO_x emissions decreased by 70% and 28% separately from 2013 to 2020, comparable to a 69% and 35% decrease in OMI column densities over eastern China and a 78% and 30% decrease in ground observations over 74 cities for the same time. The estimated emission trends are closer to satellite retrieval trend for both SO_2 and NO_x in the two periods. For example, SO_2 emissions decreased by 56% and 32% for Phase I and II respectively, consistent with 57% and 28% decrease in SO_2 columns. Discrepancies exist in the emission trend and the surface observation trend. For example, estimated emission reduction trend of 32% and 10% for SO_2 and NO_x in Phase II are significantly lower than the decrease in surface observations with 47% for SO_2 and 23% for NO_2.

Different trends between national emissions and observations could be attributed to the following reasons: (1) Variations in meteorology, atmospheric transport, and chemical reactions may contribute to the variations in air pollution concentrations in addition to emission changes. Previous studies have found non-negligible influence from the above factor on the interannual variation of column density and surface observations. (2) Satellite-based observations are more sensitive to high-stack emission sources (e.g., power plants), while ground-based observations are more influenced by surface emission sources, which might partially explain the closer relationship between OMI NO_2 and NO_x emissions when power emissions were substantially mitigated. (3) The satellite column densities were averaged over eastern China where anthropogenic emissions are dominated, while the ground observations were averaged over 74 cities over China, which are cities located in the most polluted region in China (i.e., the BTH region) and capital cities in each province. (4) Uncertainties in the ground- and satellite-based observations might also contribute to the discrepancies. Although in-situ observations are generally considered more accurate, surface NO_2 concentrations obtained from national monitoring networks may overestimate actual concentrations due to the use of chemiluminescence measurements[8]. Satellite retrievals are usually subject to higher uncertainties[9,10], mainly arising during the calculation of air mass factors. (5) The incomplete knowledge of underlying data causes large uncertainties in the bottom-up emission estimates. Usually, uncertainties are smaller for species whose emissions are dominant by stationary sources (e.g., SO_2 and NO_x) but larger for species whose emissions are mainly contributed by scattered emitting sources (e.g., NMVOC and BC)[11].

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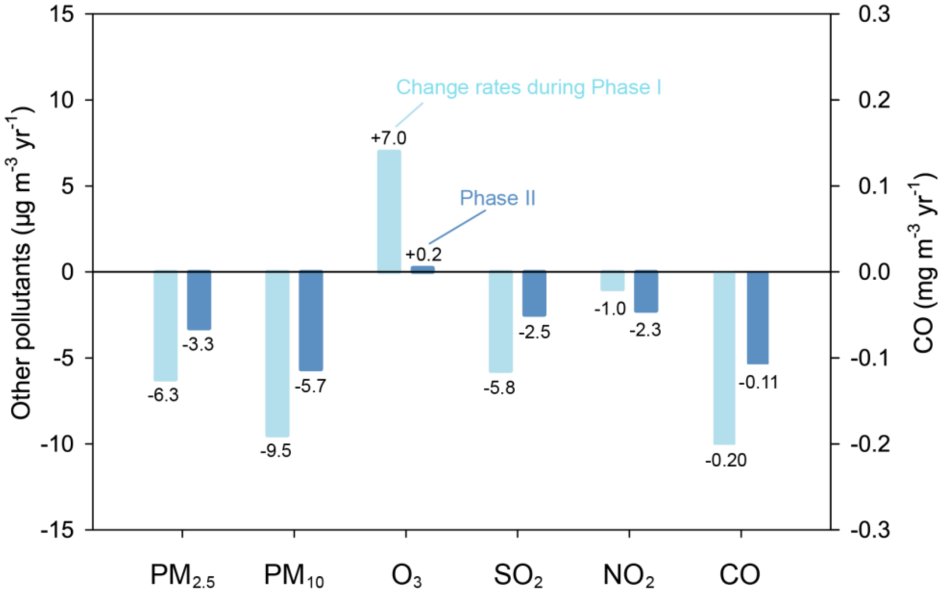
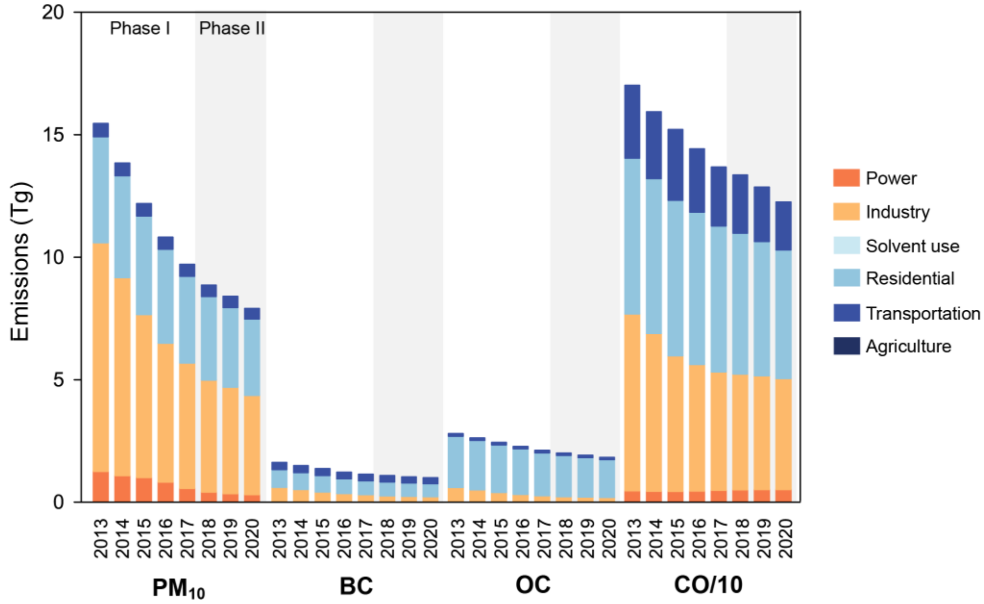
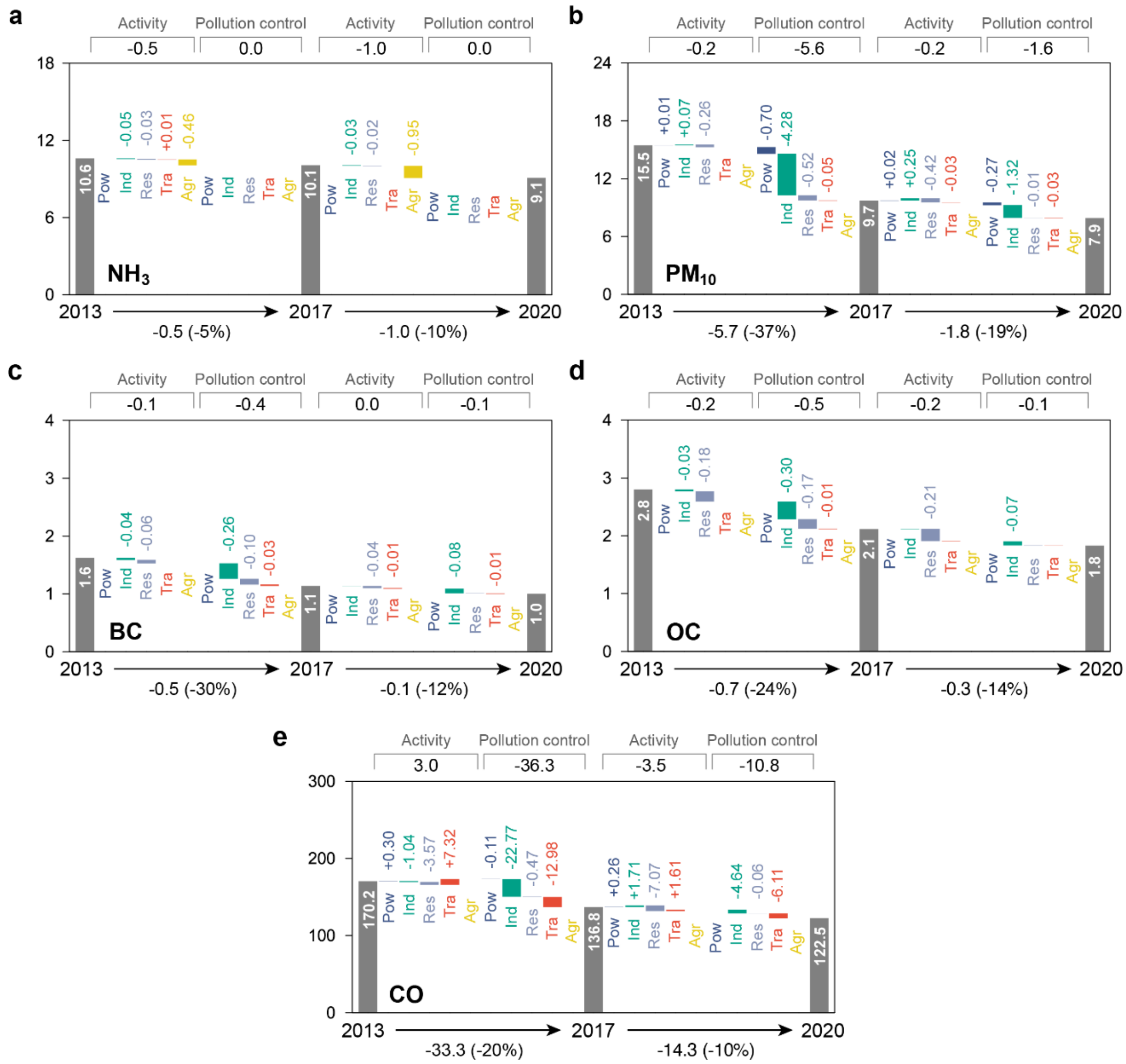
Supplementary Figures:



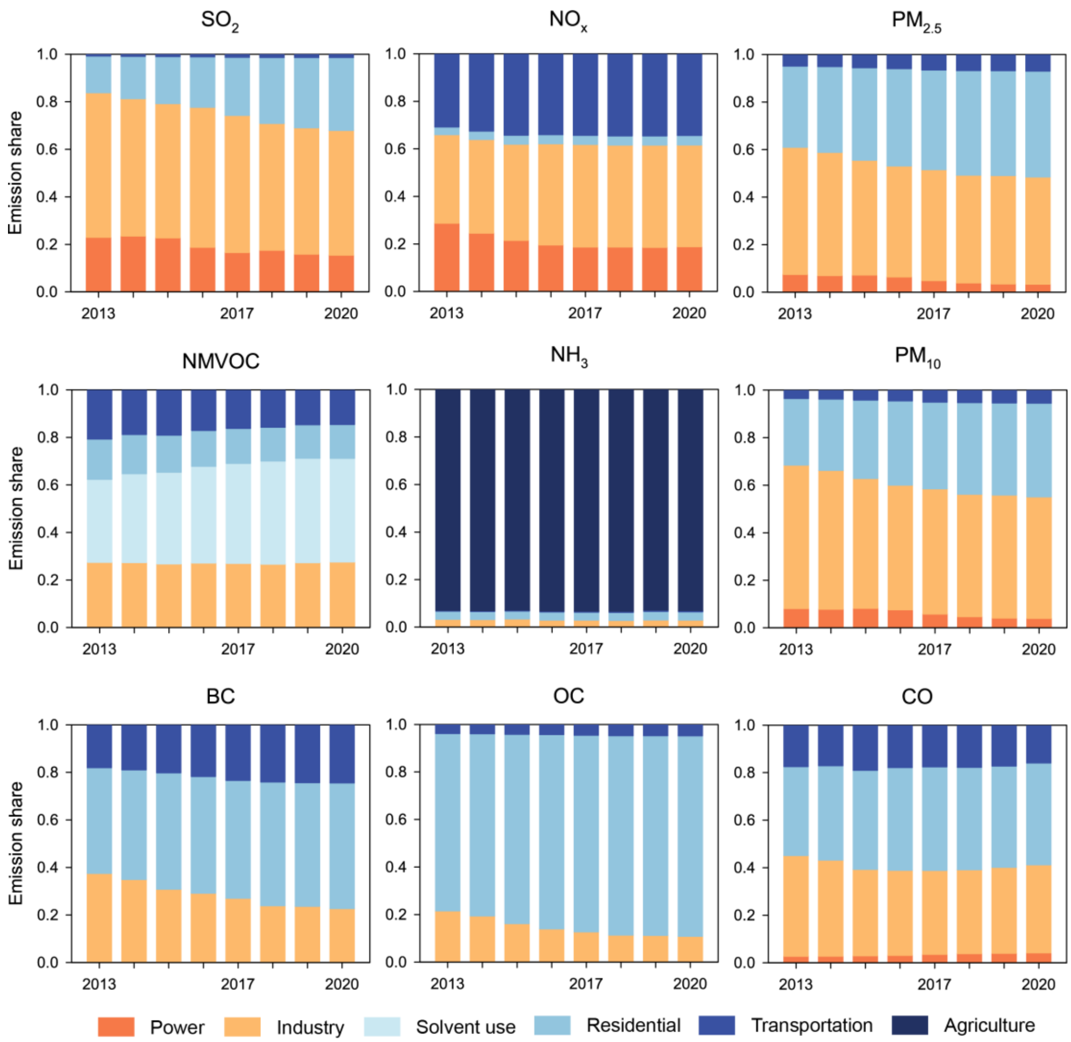
Supplementary Figure 1 | Observed annual concentration change rates of six major air pollutants during the two phases of clean air actions. The concentrations are averaged over 74 key Chinese cities that have continuous observations since the year 2013.



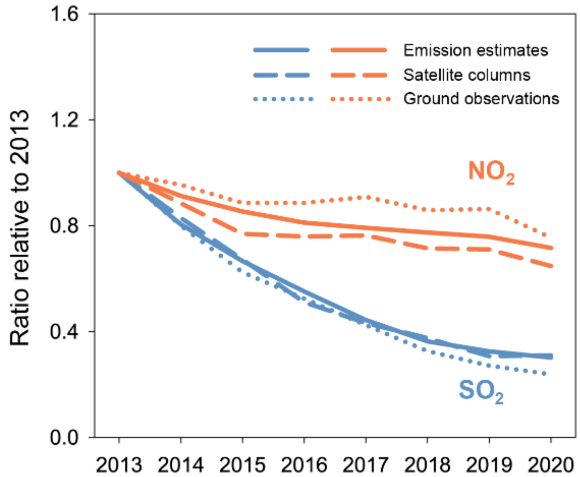
Supplementary Figure 2 | Anthropogenic emissions by sector in China during 2013–2020. This is a supplement of Figure 2, which presents the emission trends for PM_{10}, BC, OC, and CO.



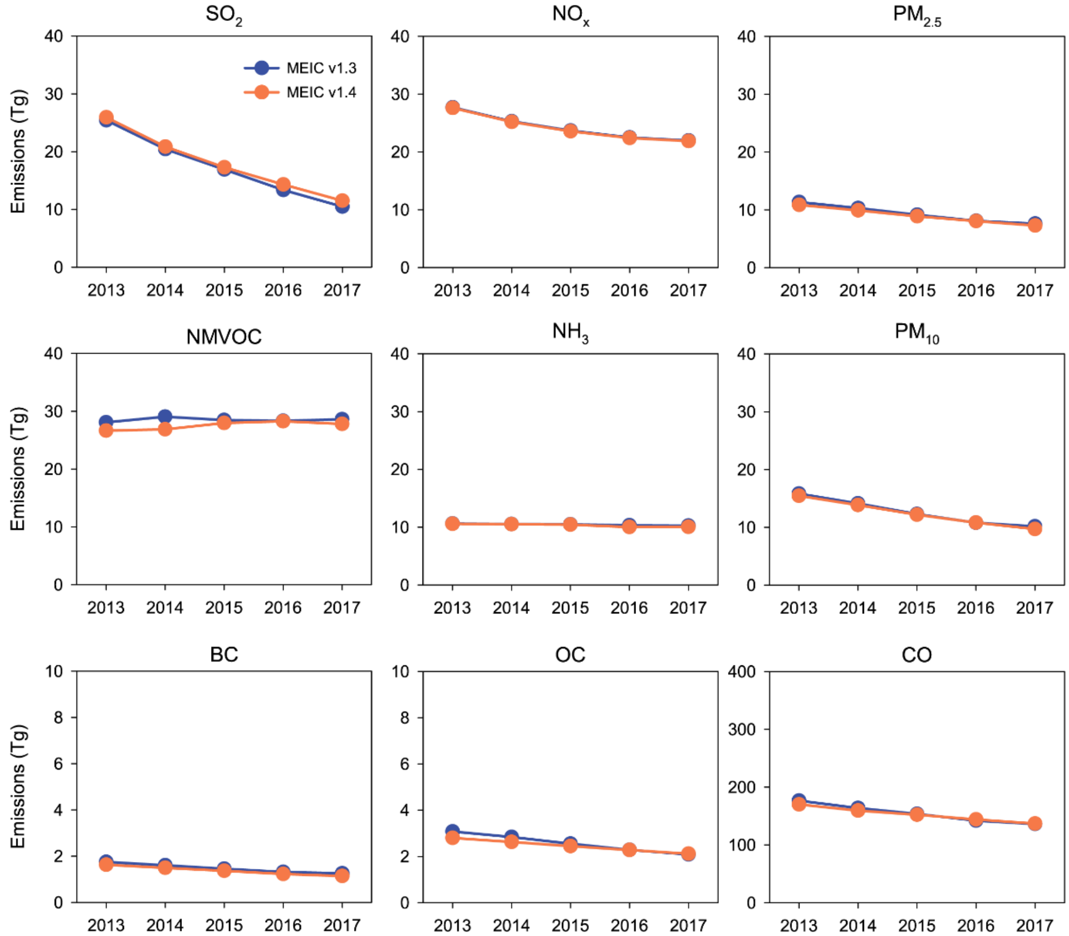
Supplementary Figure 3 | Drivers of emission changes for major air pollutants in China 2013–2020. Drivers of national emission changes in (a) NH_3, (b) PM_{10}, (c) BC, (d) OC, and (e) CO. For each pollutant, the changes in emissions are decomposed into drivers of activity rates and pollution control by sector for Phase I and Phase II of the clean air actions.



Supplementary Figure 4 | Changes in emission shares across source sectors from 2013 to 2020. Emissions are divided into six sectors: power, industry, solvent use, residential, transportation, and agriculture.



Supplementary Figure 5 | Emission trends compared with satellite- and ground-based observations. The 2013–2020 trends in SO_2 (blue solid curve) and NO_x (orange solid curve) emissions are compared with OMI SO_2 (blue dashed curve) and NO_2 (orange dashed curve) tropospheric columns for eastern China (29°N–41°N, 108°E–123°E), respectively. The 2013–2020 trends in ground-based observations of SO_2 (blue dotted curve) and NO_2 (orange dotted curve) are also presented. Data are normalized by their corresponding value in 2013.



Supplementary Figure 6 | Comparison of emission estimates between MEIC v1.3 and MEIC v1.4 during 2013–2017.

Supplementary Tables:

Supplementary Table 1 | Anthropogenic emissions of major air pollutants by sector in China from 2013 to 2020 (unit: Tg).

Species	Sectors	2013	2014	2015	2016	2017	2018	2019	2020
SO_2	Power	6.0	4.9	3.9	2.7	1.9	1.7	1.4	1.2
	Industry	15.8	12.1	9.8	8.4	6.7	5.0	4.5	4.1
	Solvent use	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Residential	4.0	3.7	3.4	3.0	2.8	2.6	2.5	2.4
	Transportation	0.2	0.2	0.2	0.2	0.1	0.1	0.1	0.1
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	25.9	20.9	17.3	14.3	11.5	9.4	8.4	7.8
NO_x	Power	8.0	6.2	5.1	4.4	4.1	4.0	3.9	3.7
	Industry	10.3	10.0	9.5	9.5	9.5	9.2	9.0	8.5
	Solvent use	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Residential	0.9	0.9	0.9	0.9	0.8	0.8	0.8	0.8
	Transportation	8.5	8.2	8.1	7.6	7.5	7.4	7.2	6.8
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	27.6	25.2	23.6	22.4	21.9	21.4	20.9	19.8
$PM_{2.5}$	Power	0.8	0.7	0.6	0.5	0.4	0.3	0.2	0.2
	Industry	5.8	5.1	4.3	3.8	3.4	3.0	2.9	2.7
	Solvent use	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Residential	3.7	3.6	3.5	3.3	3.1	3.0	2.8	2.7
	Transportation	0.5	0.5	0.5	0.5	0.5	0.5	0.4	0.4
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	10.9	9.9	8.9	8.1	7.3	6.7	6.4	6.1
NMVOC	Power	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	Industry	7.3	7.3	7.5	7.6	7.5	7.4	7.3	7.0
	Solvent use	9.3	10.0	10.8	11.5	11.7	12.1	11.9	11.2
	Residential	4.5	4.4	4.4	4.3	4.1	4.0	3.8	3.7
	Transportation	5.5	5.1	5.3	4.8	4.5	4.4	4.0	3.7
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	26.7	26.9	28.0	28.3	27.8	27.8	27.1	25.6
NH_3	Power	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Industry	0.4	0.3	0.4	0.3	0.3	0.3	0.3	0.3
	Solvent use	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Residential	0.4	0.4	0.4	0.3	0.3	0.3	0.3	0.3
	Transportation	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
	Agriculture	9.9	9.8	9.7	9.4	9.4	9.0	8.3	8.4
	Total	10.6	10.5	10.5	10.0	10.1	9.6	9.0	9.1
PM_{10}	Power	1.3	1.1	1.0	0.8	0.6	0.4	0.3	0.3
	Industry	9.3	8.1	6.7	5.7	5.1	4.6	4.3	4.1
	Solvent use	0.5	0.5	0.5	0.5	0.5	0.5	0.4	0.4
	Residential	4.3	4.2	4.0	3.8	3.5	3.4	3.3	3.1
	Transportation	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	15.5	13.8	12.2	10.8	9.7	8.9	8.4	7.9
BC	Power	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Industry	0.6	0.5	0.4	0.4	0.3	0.3	0.2	0.2
	Solvent use	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.2
	Residential	0.7	0.7	0.7	0.6	0.6	0.6	0.5	0.5
	Transportation	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	1.6	1.5	1.4	1.2	1.1	1.1	1.0	1.0
OC	Power	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Industry	0.6	0.5	0.4	0.3	0.3	0.2	0.2	0.2
	Solvent use	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	Residential	2.1	2.0	1.9	1.9	1.7	1.7	1.6	1.5
	Transportation	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	2.8	2.6	2.4	2.3	2.1	2.0	1.9	1.8
CO	Power	4.7	4.5	4.5	4.6	4.9	5.1	5.2	5.1
	Industry	72.1	64.4	55.3	51.6	48.3	47.2	46.5	45.4
	Solvent use	29.7	27.2	28.9	25.8	24.0	23.7	22.2	19.5
	Residential	63.6	63.2	63.5	62.2	59.6	57.5	54.8	52.5
	Transportation	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Agriculture	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	170.2	159.4	152.1	144.2	136.8	133.6	128.6	122.5

Supplementary Table 2 | Comparison of national and sectoral emission trends between MEIC v1.3 and v1.4.

Species		Version	Emission changes between 2013 and 2017						
			Pow	Ind	Tra	Res	Agr	Sol	Total
SO_2	Absolute change/Tg	v1.3	-4.1	-9.9	0.0	-1.0	-	-	-14.9
		v1.4	-4.0	-9.1	0.0	-1.2	-	-	-14.4
	Relative change	v1.3	-69%	-62%	15%	-29%	-	-	-59%
		v1.4	-68%	-58%	-26%	-29%	-	-	-56%
NO_x	Absolute change/Tg	v1.3	-3.8	-1.1	-0.8	-0.1	-	-	-5.7
		v1.4	-3.9	-0.8	-1.0	-0.1	-	-	-5.7
	Relative change	v1.3	-47%	-10%	-9%	-13%	-	-	-21%
		v1.4	-49%	-8%	-12%	-7%	-	-	-21%
$PM_{2.5}$	Absolute change/Tg	v1.3	-0.3	-2.3	0.0	-1.2	-	-	-3.7
		v1.4	-0.5	-2.4	-0.1	-0.7	-	-	-3.6
	Relative change	v1.3	-31%	-40%	3%	-28%	-	-	-33%
		v1.4	-56%	-41%	-10%	-18%	-	-	-33%
NMVOC	Absolute change/Tg	v1.3	0.0	0.6	-0.8	-1.2	-	1.9	0.5
		v1.4	0.0	0.2	-1.0	-0.4	-	2.4	1.2
	Relative change	v1.3	7%	7%	-14%	-25%	-	22%	2%
		v1.4	8%	3%	-18%	-9%	-	26%	4%
NH_3	Absolute change/Tg	v1.3	-	0.0	0.0	-0.1	-0.2	-	-0.4
		v1.4	-	-0.1	0.0	0.0	-0.5	-	-0.5
	Relative change	v1.3	-	-14%	25%	-23%	-2%	-	-3%
		v1.4	-	-15%	27%	-8%	-5%	-	-5%
PM_{10}	Absolute change/Tg	v1.3	-0.3	-4.1	0.0	-1.3	-	-	-5.7
		v1.4	-0.7	-4.2	-0.1	-0.8	-	-	-5.7
	Relative change	v1.3	-24%	-44%	3%	-27%	-	-	-36%
		v1.4	-55%	-45%	-10%	-18%	-	-	-37%
BC	Absolute change/Tg	v1.3	0.0	-0.3	0.0	-0.2	-	-	-0.5
		v1.4	0.0	-0.3	0.0	-0.2	-	-	-0.5
	Relative change	v1.3	-31%	-47%	4%	-25%	-	-	-28%
		v1.4	-56%	-49%	-9%	-22%	-	-	-30%
OC	Absolute change/Tg	v1.3	0.0	-0.3	0.0	-0.7	-	-	-1.0
		v1.4	0.0	-0.3	0.0	-0.3	-	-	-0.7
	Relative change	v1.3	-34%	-53%	2%	-29%	-	-	-32%
		v1.4	-61%	-55%	-10%	-16%	-	-	-24%
CO	Absolute change/Tg	v1.3	0.1	-23.7	-4.6	-12.3	-	-	-40.4
		v1.4	0.2	-23.8	-5.7	-4.0	-	-	-33.3
	Relative change	v1.3	3%	-33%	-15%	-18%	-	-	-23%
		v1.4	4%	-33%	-19%	-6%	-	-	-20%