Assessment of strict autumn-winter emission controls on air quality in the Beijing-Tianjin-Hebei region

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Abstract. Strict seasonal emission controls are a popular measure in China for addressing severe air pollution, in particular fine particulate matter (PM\textsubscript{2.5}). Here we evaluate the efficacy of these measures, with a particular focus on the strict emission controls imposed on pollution sources in 28 cities in and around the Beijing-Tianjin-Hebei region (BTH) in autumn-winter 2017/2018. For this we use the GEOS-Chem chemical transport model and air pollutant measurements from the national and Beijing local monitoring networks, after evaluating the network data with independent measurements and correcting large biases in the bottom-up emissions inventory. The network measurements are temporally consistent ($r > 0.9$ for PM\textsubscript{2.5} and $r > 0.7$ for gases) with the independent measurements, though with systematic differences of 5-17\% for nitrogen dioxide (NO\textsubscript{2}) and 16-28\% for carbon monoxide (CO). The average decrease in monitoring network PM\textsubscript{2.5} in BTH in autumn-winter 2017/2018 relative to the previous year is 27\%, declining from 103 to 75 $\mu$g m\textsuperscript{-3}. The regional decline in PM\textsubscript{2.5} in the model is 20\%, exceeding the regional target of 15\%. According to the model, pollution control measures led to decline in PM\textsubscript{2.5} precursor emissions of 0.27 Tg NO\textsubscript{x} (as NO), 0.66 Tg sulfur dioxide (SO\textsubscript{2}), 70 Gg organic carbon (OC), and 50 Gg black carbon (BC). We find though that these alone only lead to an 8\% decline in PM\textsubscript{2.5} and that interannual variability in meteorology accounts for more than half (57\%) of the decline. This demonstrates that year-on-year comparisons are misleading for assessing the efficacy of air pollution measures and should be taken into consideration when extending such measures beyond BTH.

1 Introduction

Strict seasonal emission controls are an increasingly popular measure to reduce severe air pollution in China, in particular elevated concentrations of fine particles, or PM\textsubscript{2.5}, in autumn-winter. The Chinese government improved such measures in 28 cities in and around the Beijing-Tianjin-Hebei region (BTH) (so-called “2+26” cities) in

northern China in autumn and winter 2017/2018. Targets were set to reduce regional mean PM$_{2.5}$ by 15% and
city-specific PM$_{2.5}$ by 10-25% relative to the previous year (MEE, 2017). BTH experiences severe air pollution
in China despite substantial decline in PM$_{2.5}$ of ~40% from 2013 to 2017 from enacting emission controls as part
of the 5-year Action Plans (Zhang et al., 2019). Annual mean PM$_{2.5}$ in BTH in 2017 was 64 µg m$^{-3}$ (Wang et al.,
2019); far greater than the national standard of 35 µg m$^{-3}$ (MEE, 2012) and the World Health Organization (WHO)
guideline of 10 µg m$^{-3}$. Severe pollution days (defined as days with 24-hour mean PM$_{2.5}$ ≥ 150 µg m$^{-3}$), mostly in
autumn and winter, have declined in frequency, but still occurred in 78 days in 2016. Down from 122 days in
2013 (Li et al., 2019a). Severe PM$_{2.5}$ pollution is due to a combination of large primary emissions of particles and
gas-phase precursors from multiple sources (Zhang et al., 2018), very active heterogeneous chemistry enhancing
formation of secondary inorganic and organic aerosols (Huang et al., 2014), and accumulation of pollution due to
meteorological conditions such as low windspeeds, shallow planetary boundary layer and high relative humidity
(RH) (An et al., 2019; Bei et al., 2020; Le et al., 2020; Wu et al., 2019).

Dominant local PM$_{2.5}$ sources in BTH in autumn-winter include sustained contributions from the energy sector
and road traffic (Tong et al., 2020), and seasonal contributions from industrial and residential combustion of coal
and other solid fuels (Ma et al., 2017; Yun et al., 2020), and widespread burning of crop residue (Li et al., 2020c).
Mitigation measures in China have led to a nation-wide decrease in emissions of the primary PM$_{2.5}$ components
black carbon (BC) and organic carbon (OC) of 28 % for BC and 32 % for OC from 2013 to 2017 (Zheng et al.,
2018). Emissions of prominent gas-phase PM$_{2.5}$ precursors such as nitrogen oxides (NO$_x$ = NO + NO$_2$) and sulfur
dioxide (SO$_2$) have declined by 21% for NO$_x$ and 59% for SO$_2$ over the same time period. Trends in other PM$_{2.5}$
precursors are less certain. Emissions of ammonia (NH$_3$), mostly from agriculture, are likely to have increased or
remained constant (Zheng et al., 2018). Similar to NH$_3$, emissions of non-methane volatile organic compounds
(NMVOCs), mostly from industrial activity and solvent use, are either stable or increasing (Li et al., 2019c; Liu et al.,
2018b; Zhang et al., 2017a; Zheng et al., 2018). Non-local sources also make a substantial contribution to
PM$_{2.5}$ in BTH throughout the year. Dong et al. (2020) used a regional air quality model to estimate that regional
transport of non-local PM$_{2.5}$ accounted for 33-68% of total monthly mean PM$_{2.5}$ in BTH in 2017.

Many mitigation measures were implemented in autumn-winter 2017/2018 to reduce PM$_{2.5}$ in the 28 cities in and
around BTH. These are detailed in the “Air Pollution Action Plan in Autumn and Winter of 2017-2018” for the
Beijing-Tianjin-Hebei Region and its Surrounding Areas” report by the Chinese Ministry of Ecology and
Environment (MEE) (MEE, 2017). Briefly, these include a sector-wide cap on total consumption of coal, phase-
out of small inefficient and outdated industrial coal-fired boilers, reduction in production capacity of heavy
industries such as iron and cement, switching from coal to cleaner fuels in homes, and mandated controls on
construction site fugitive dust emissions. Other short-term and reactionary measures included shutdown of
intensive industries and construction sites throughout the emission control period and instantaneous shutdown of
additional industrial plants in response to forecasts of elevated PM$_{2.5}$. Tougher emission standards and higher
quality vehicular fuel were imposed on on-road vehicles. Agricultural residue burning was banned and strictly
enforced, and installation of emission control technologies were mandated for all large emitters of industrial
NMVOCs. The MEE used national network observations of PM$_{2.5}$ to determine that regional reduction targets
were achieved in BTH and that only 3 of the 28 cities did not meet their city-specific targets (MEE, 2018).
As demonstrated by studies assessing changes in air quality during the lockdown period in response to the SARS-CoV-2 pandemic, variability in meteorology is also a large contributor to fluctuations in air quality in BTH (Le et al., 2020; Shi et al., 2021). This necessitates that assessment of the efficacy of such measures includes detailed understanding of the contribution of strict emission controls and other factors like meteorology. A recent study by Zhang et al. (2021) assessed city-scale changes in air quality in each of the 28 cities using the high-resolution Community Multi-Scale Air Quality model coupled to the Weather Research and Forecasting Model for meteorology (CMAQ-WRF). CMAQ-WRF was driven with a national bottom-up inventory for the year preceding the emission controls and a regional bottom-up emission inventory for the year of the emission control period. They determined that the contribution of emission controls to the decrease in simulated PM$_{2.5}$ in each city ranged from 2% to 82% and that meteorology was often a dominant contributor, ranging from 18% to 98%. Here we take a regional perspective, after correcting for large biases in the bottom-up emission inventories for BTH with the China national and Beijing regional monitoring network observations which we also assess against independent measurements. We apply the corrected inventory to the GEOS-Chem chemical transport model (CTM) to determine regional-scale emission reductions resulting from strict mitigation measures in autumn-winter 2017/2018 and the contribution of these and meteorology to improved regional air quality, as such measures are now widely adopted in China.

2 Air pollutant concentration changes detected with the national and local monitoring networks

Ambient monitoring of PM$_{2.5}$ and trace gases in BTH includes reference monitors from the China National Environmental Monitoring Network (CNEMN) and the local Beijing Municipal Environmental Monitoring Network (BJMEMN) (Zhang et al., 2020). We use observations of hourly SO$_2$, NO$_2$, CO, and PM$_{2.5}$ for the autumn-winter emission reduction period (2017/2018) and the preceding year (2016/2017) at sites operational in both years: 402 for CNEMN and 35 for BJMEMN. Data from both networks are from the Sina Air Quality Data Platform (http://beijingair.sinaapp.com/; last accessed 17 October 2020, now hosted at https://aqicn.org/). Data from both networks have been extensively used to quantify changes in surface air pollution (Li et al., 2019b; Silver et al., 2018; Wan et al., 2021; Wang et al., 2014; Zhai et al., 2019), though independent evaluation of the measurements is limited. A previous study used statistical techniques and intercomparison of air pollutant measurements to determine that outliers make only a small contribution (≤ 1%) to measurements of air pollutants relevant to this work (Wu et al., 2018).

We assess CNEMN and BJMEMN measurements against PM$_{2.5}$ from the US Embassy in Beijing and PM$_{2.5}$ and trace gas (SO$_2$, NO$_2$, and CO) measurements from the winter portion of the intensive Atmospheric Pollution & Human Health in a Chinese Megacity (APHH) campaign, hereafter referred to as APHH. The APHH campaign included a comprehensive suite of aerosol and gas-phase measurements from the 325-m tower at the urban Institute of Atmospheric Physics (IAP) measurement site and a few air quality measurements at a rural site (Pinggu) located ~60 km from the Beijing city centre (Shi et al., 2019). APHH data are from the National Environmental Research Council (NERC) Centre for Environmental Data Archive (CEDA) (Fleming et al., 2017). US Embassy PM$_{2.5}$ measurements in Beijing, obtained with US EPA measurement and quality control protocols (Martini et al., 2015), are from the US Department of State Air Quality Monitoring Program (http://www.stateair.net/; last
accessed 17 October 2020) for autumn-winter 2016/2017 and from the OpenAQ data portal (http://www.openaq.org/; last accessed 17 October 2020) for autumn-winter 2017/2018. We use APHH data for November-December 2016 and US Embassy PM$_{2.5}$ for October 2016-March 2017 and October 2017-March 2018. These are compared to the nearest national and local monitoring network sites. For APHH, these are the CNEMN Aotizhongxin site (39.98°N, 116.40°E) and the BJMEMN Xizhimenbei site (39.95°N, 116.35°E), each located ~3 km from the APHH urban site (39.97°N 116.37°E). For the US Embassy, these are the CNEMN site Nongzhanguan (39.94°N, 116.46°E) and the BJMEMN site Dongsihuan (39.94°N, 116.48°E), each ~1 km from the US Embassy (39.95°N, 116.47°E).

Figure 1 compares hourly CNEMN and BJMEMN PM$_{2.5}$ to APHH and US Embassy PM$_{2.5}$, PM$_{2.5}$ from both CNEMN and BJMEMN are temporally consistent with APHH and US Embassy PM$_{2.5}$ ($r \geq 0.96$). The surface monitoring networks also reproduce the variance in hourly PM$_{2.5}$ (Slopes of 1.0-1.1). Network sites are systematically higher than APHH by 10% for CNEMN and 17% for BJMEMN, though compared to US Embassy site PM$_{2.5}$ the difference is small, ranging from negligible (0.2% less) to 6.4% more than US Embassy PM$_{2.5}$. In general, the BJMEMN measurements are 6-17% more than APHH, likely due to spatial variability in local emissions. The decline in PM$_{2.5}$ in the control period relative to the previous year at these sites is 43% according to US Embassy PM$_{2.5}$, decreasing from 97 µg m$^{-3}$ to 55 µg m$^{-3}$. A similar decline is obtained with the nearby BJMEMN (43% decline) and CNEMN (42% decline) sites shown in Fig. 1.

Figure 2 compares hourly trace gas measurements from CNEMN, BJMEMN and APHH. The CNEMN and BJMEMN trace gas instruments include chemiluminescence for NO$_{2}$, UV fluorescence for SO$_{2}$, and IR absorption for CO. Though CO is not a precursor of PM$_{2.5}$, its abundance affects the oxidative potential of the atmosphere and also offers a means to indirectly assess PM$_{2.5}$ precursor emissions of NMVOCs that oxidize to form CO. Most measurements from the local and national networks, with the exception of 31% of the CNEMN SO$_{2}$ data and 16% of the BJMEMN SO$_{2}$ data, are above the instrument detection limit (indicated in Fig. 2). The surface network is temporally consistent with APHH ($r > 0.7$ for all species). CNEMN NO$_{2}$ is <10% more than NO$_{2}$ from APHH, likely due to susceptibility of the monitoring network instruments to interference from decomposition of NO$_{x}$ reservoir compounds to NO$_{2}$ (Dunlea et al., 2007; Reed et al., 2016). Shah et al. (2020) estimate a positive bias of ~6% due to this interference. Differences in CNEMN SO$_{2}$ (19% less than APHH) and CO (16% more than APHH) are large and reflect differences in variance (CNEMN vs APHH SO$_{2}$ slope of 0.8, CNEMN vs APHH CO slope of 1.4). BJMEMN exceeds APHH by 17-28% for all trace gases.

To aid interpretation of the differences between the network sites and independent measurements, we also assess consistency between CNEMN and BJEMN for the sites shown in Figs. 1 and 2. These are ~5 km apart and the BJEMN site is closer to a heavily trafficked ring road than the CNEMN site. The sites are strongly correlated for PM$_{2.5}$ ($r = 0.97$) and all trace gases ($r = 0.89-0.92$), but CNEMN is less than BJEMN by 11% for NO$_{2}$, 33% for SO$_{2}$, 9% for CO, and 6% for PM$_{2.5}$ due to the lower relative influence of road traffic sources. This is consistent with the relatively large positive differences between BJEMN and the independent measurements in Figs. 1 and 2.
Both networks also measure ozone, but we do not consider this here, as ozone pollution is most severe in spring and summer in China (Yang et al., 2020). We estimate mean ozone of 15-19 µg m⁻³ at the two monitoring network sites used in Fig. 2. This can be compared to the summer mean ozone air quality metric, mean maximum daily 8-h average ozone, of 120-160 µg m⁻³ in northern China (Li et al., 2019b).

We show in Fig. 3 the spatial distribution of the network observed changes in air pollutant concentrations in and around BTH in the control period (autumn-winter 2017/2018) relative to the preceding year (autumn-winter 2016/2017). In what follows, we refer to these time periods as AW2017 for autumn-winter 2017/2018 and AW2016 for autumn-winter 2016/2017. Only CNEMN and BJMEMN sites that are operational in both periods are used. These include 164 sites within the control domain (region shaded grey in Fig. 3) and 273 sites in the surrounding area. The decline in air pollutant concentrations in AW2017 relative to AW2016 at sites within the emission control region is 16% for NO₂, 44% for SO₂, 31% for CO, and 29% for PM₂.5, surpassing the 15% PM₂.5 reduction target set for BTH. Surface concentrations of ozone (not shown) increase by 19% in response to decline in NOₓ. Even with this increase, ozone is still substantially lower than in spring and summer (Liu et al., 2018a). Smaller reductions of 0.5% for NO₂, 31% for SO₂, 13% for CO, and 10% for PM₂.5 occur in the surrounding area.

In the southeast portion of the domain shown in Fig. 3, both NO₂ and PM₂.5 increase by 5-8%. Fang et al. (2019) reported an increase in emissions from industries in the non-control area that in Fig. 3 appear to offset air quality improvements that would be expected from decline in influence of pollution from BTH.

### 3 BTH air pollution and emissions for the year preceding emission controls

We use the GEOS-Chem model (version 12.0.0; https://doi.org/10.5281/zenodo.1343547) with the evaluated surface network measurements to constrain precursor emissions of PM₂.5 in and around BTH. The model is nested over East Asia (11°S-55°N, 60-150°E) at a horizontal resolution of 0.5° × 0.625° (latitude × longitude). The model is driven with assimilated meteorology from the NASA Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2) updated hourly for 2D fields and every 3 hours for 3D fields. Dynamic (3-hourly) boundary conditions are from a global simulation at 4° × 5°. Monthly anthropogenic emissions for China in AW2016 are from the regional bottom-up Multi-resolution Emission Inventory for China (MEIC) (http://www.meicmodel.org; last accessed 4 March 2020) available for 2000-2017 at 0.5° × 0.625°. MEIC includes emissions of SO₂, NOₓ, CO, NMVOCs, NH₃, and primary particles from ~700 anthropogenic sources (Zheng et al., 2018; Li et al., 2017). In its implementation in GEOS-Chem, MEIC emissions are lumped into five sectors: industry, power plants, transportation, agriculture and residential. Primary particles are emitted as hydrophobic and hydrophilic BC and OC, and speciated NMVOCs are mapped to those in GEOS-Chem using the NMVOCs species mapping tables in Li et al. (2014).

The model includes detailed coupled gas- and aerosol-phase chemistry to represent the formation and loss of PM₂.5. Individual aerosol components are modelled as externally mixed. These include sulfate, nitrate, ammonium (Park et al., 2004; Wang et al., 2013), OC (Heald et al., 2006), BC (Li et al., 2016), dust (Fairlie et al., 2007), and sea salt (Jaegle et al., 2011). Formation of secondary sulfate-nitrate-ammonium aerosols are computed with ISORROPIA-II (Fountoukis and Nenes, 2007). Physical loss processes include dry and wet deposition (Amos et al., 2012; Liu et al., 2001; Wang et al., 1998). We implement a revised treatment of wet scavenging described and
first implemented in GEOS-Chem by Luo et al. (2019). This replaces fixed values of in-cloud condensation water with dynamic values from MERRA-2. This leads to more rapid wet deposition rates and addresses a positive bias in modelled nitrate and ammonium, in particular in winter, when compared to surface observations in China, Europe, and the US (Luo et al., 2020). We sample the model in AW2016 and AW2017 following two months spin-up before each period of interest for chemical initialization.

We find from initial comparison of the model to the surface observations that the model in AW2016 is considerably less than observed NO₂ (by 48%), SO₂ (by 42%), and CO (by 57%) over the entire domain shown in Fig. 3. We attribute this to an underestimate in precursor emissions of these in the MEIC. Previous studies reported that MEIC trends in NO₂, SO₂, and CO emissions are consistent with trends in satellite observations of column densities and weather-normalised surface measurements (Vu et al., 2019; Zheng et al., 2018), but studies assessing and identifying similar underestimates to ours are limited to very local assessment of the inventory. Squires et al. (2020) determined that MEIC NO₂ and CO emissions are overestimated at the urban APHH site from comparison to fluxes calculated using the eddy-covariance method, though their comparison was for different years (measurements in 2016, MEIC in 2013). We calculate scale factors to apply to the MEIC based on our initial comparison to the network site measurements. These include spatially uniform scale factors of 1.5 applied to NO₂ emissions and 2.4 applied to CO emissions across the whole domain shown in Fig. 3. Spatially variable scale factors of 2.1-6.8 are applied to seven grid squares for the MEIC SO₂ emissions. These are concentrated in Shanxi province west of BTH, a region with large coal-fired power plants (Xie et al., 2018). These grids account for the majority of the domain average 42% underestimate in modelled SO₂ concentrations. Local enhancements in CO can include primary emissions and secondary contributions from oxidation of NMVOCs.

Emission inventory estimates of NMVOCs are subject to large uncertainties, mainly due to poorly quantified industrial emissions (Li et al., 2017) and lack of reliable data for scattered areal sources such as residential coal burning (Li et al., 2019c; Peng et al., 2019; Shi et al., 2020). We find though that modelled CO is relatively unaffected by NMVOCs emissions. A sensitivity simulation with MEIC NMVOCs emissions increased by 50% only increases surface CO concentrations by 0.2%. Given this, we attribute the model underestimate in CO surface concentrations to the emissions. We do not adjust MEIC primary emissions of BC and OC, due to limited constraints on these from the observations. APHH eddy covariance fluxes of BC obtained in winter suggest a large overestimate (59 times) in MEIC BC emissions (Joshi et al., 2021), though this may be due to very local influence of traffic that is diluted at the resolution of the MEIC (Joshi et al., 2021).

Figure 4 compares GEOS-Chem and monitoring network air pollutant concentrations for BTH and the surrounding area after applying scale factors to MEIC emissions of SO₂, NO₂, and CO. Emissions scaling of NO₂ improves the modelled variance in NO₂. The regression slope increases from 0.86 (not shown) to 0.93 (Fig. 4). There is still an underestimate in modelled background NO₂ (intercept = -12.8 µg m⁻³) that leads to a model NO₂ normalized mean bias (NMB) of -33%. The monitoring network includes sites close to busy roads that may be influenced by local traffic emissions that would be diluted at the 50-67 km resolution of the model. The BJMEMN sites are categorised by location and we find that excluding roadside sites leads to a 5-8% decrease in mean BJMEMN NO₂ (2-8% decrease for the other pollutants) and would only partially resolve the remaining underestimate in modelled NO₂ in Fig. 4. Other factors that could contribute to the remaining discrepancy include...
short atmospheric lifetime of NO2, relatively coarse model resolution, and a positive 5-17% bias in the monitoring network measurements (Fig. 2). Any further increases in MEIC NOx emissions would worsen the model NMB in PM2.5 of 15%, as nitrate from oxidation of NOx is the dominant component of PM2.5 (29% in AW2016 for the entire domain in Fig. 3, according to GEOS-Chem).

Emissions scaling of SO2 improves spatial consistency for concentrations of SO2 from $r = 0.54$ (not shown) to $r = 0.90$ (Fig. 4). There is still a model bias -29% mostly due to 2 grids in Shanxi province with observed SO2 of 161-200 μg m$^{-3}$ and modelled SO2 of 109-122 μg m$^{-3}$ (Fig. 4). The model underestimate in CO decreases from -57% to -14% and the remaining model bias may be due to a 16-28% positive bias in the monitoring network (Fig. 2). The improvement in spatial correlation for CO is marginal, increasing from $r = 0.35$ to $r = 0.45$. Scaling MEIC emissions of SO2 and NOx increases the modelled PM2.5 NMB from 8% with the original emissions to 15% with the scaled emissions. This may be due to remaining uncertainties in MEIC emissions of BC, OC, and NH3.

Figure 5 compares observed and modelled PM2.5 composition at the urban and rural APHH sites. The model overestimates total PM2.5 by 10% at the urban site and 34% at the rural site. Components are measured with a semicontinuous analyzer for OC and BC (Han et al., 2014; Kondo et al., 2006) and ion chromatography following particle collection with a Partisol for sulfate, nitrate, and ammonium (Taiwo et al., 2014; Xu et al., 2021). OC is compared instead of organic aerosol (OA), due to uncertainties in conversion of OC to OA (Xing et al., 2013). According to the observations, OC is the dominant PM2.5 component, with a carbon mass contribution to total PM2.5 of 23% at the urban site and 33% at the rural site. The total contribution of secondary inorganic aerosols is similar (31%) at both sites and includes 9% sulfate, 13% nitrate, and 9% ammonium at the urban site and similar contributions (8% sulfate, 13% nitrate, and 10% ammonium) at the rural site. BC is 4% of total PM2.5 at both sites. The model underestimate in sulfate may in part be due to the remaining underestimate in MEIC SO2 emissions (Fig. 4) after scaling MEIC SO2 emissions. The model may also be missing key sulfate formation processes during haze events (Bloss et al., 2021; Wang et al., 2016; Wang et al., 2020), though the measurements may also be impacted by interference from hydroxymethane sulfonate (HMS) (Moch et al., 2018; Song et al., 2019). The overestimate in modelled nitrates is a common issue and has been attributed by Miao et al. (2020) to an overestimate in aerosol nitrate precursors at night exacerbated by errors in boundary layer dynamics. Despite biases in PM2.5 composition, the model reproduces day-to-day variability in 24-hour mean PM2.5 ($r = 0.66-0.71$) and its components ($r = 0.50-0.78$) at both sites.
Figure 6 shows total anthropogenic emissions of gaseous PM$_{2.5}$ precursors and primary PM$_{2.5}$ (OC and BC) for AW2016 obtained after applying scaling factors to the MEIC based on discrepancies in modelled and observed air pollutant concentrations. Total anthropogenic emissions in BTH (area shaded grey in Fig. 3) in AW2016, the year prior to the control period, are 2.4 Tg NO$_x$ as NO, 1.6 Tg SO$_2$, 150 Gg BC, 240 Gg OC, and 41 Tg CO. NMVOCs (2.0 Tg C) and NH$_3$ (0.64 Tg) are not shown in Fig. 6, as these are unchanged from AW2016 to AW2017. Though emissions of NO$_x$, CO and SO$_2$ increase in AW2016 relative to the default MEIC emissions due to our emissions scaling, there is no change in the relative contribution of different sectors. The major sector contributions include industry for NO$_x$ (44%), SO$_2$ (56%), and CO (39%), transport for NO$_x$ (34%), and residential fuel use for domestic heating and cooking for SO$_2$ (28%), CO (42%), BC (47%) and OC (79%).

4 Influence of emissions and meteorology on air quality in AW2017

To estimate the emission changes in the model due to controls implemented in BTH in AW2017, we regrid the relative changes in the surface air quality observations (Fig. 3) to a 1° × 1.25° resolution grid to achieve reasonably extensive coverage across the whole domain shown in Fig. 3. For grids without surface observations (17% of the grids in the emission control region, 51% outside it), we interpolate across nearest neighbouring grids. Anthropogenic emissions beyond the geographic limits in Fig. 3 are unchanged. Due to lack of observations of OC and BC concentrations, we initially use the relative change in total measured PM$_{2.5}$. This ranges from a decrease in AW2017 relative to AW2016 of 5% to 53% in BTH. We find with this initial approach that the model underestimates the percent reduction in PM$_{2.5}$ in AW2017 relative to AW2016. This suggests that the decline in primary PM$_{2.5}$ may be greater than the decline in total PM$_{2.5}$, corroborated by the greater decline in primary PM$_{2.5}$ emissions in the bottom-up inventories used by Zhang et al. (2021) than the decline in measured total PM$_{2.5}$ for around half of the 28 cities. This may be because the regulations mostly targeted sources that have large primary PM$_{2.5}$ emissions, such as coal combustion, industry, vehicles, fugitive dust and biomass burning (Zhang et al., 2017b; Zheng et al., 2017). We iterate to obtain BC and OC emissions scaling factors that are 1.4 times more than the percent change in total PM$_{2.5}$.

Figure 7 shows the spatial distribution of modelled and observed PM$_{2.5}$ concentrations and relative changes in PM$_{2.5}$ in and around BTH. The average decrease in observed PM$_{2.5}$ is 28% in BTH, declining from 103 μg m$^{-3}$ in AW2016 to 75 μg m$^{-3}$ in AW 2017. A similar decline is obtained with the model for grids coincident with the sites (25% decrease from 112 μg m$^{-3}$ in AW2016 to 85 μg m$^{-3}$ in AW2017). The decline in modelled PM$_{2.5}$ for all BTH grids is 20% and compared to 16% for the whole domain. In Beijing, observed PM$_{2.5}$ decreases from 96 μg m$^{-3}$ in AW2016 to 57 μg m$^{-3}$ in AW2017, a 40% reduction. The decline in the model for all 13 grids covering Beijing (39.25-41.25°N, 115.3125-117.8125°E) is more modest (33% decline from 75 μg m$^{-3}$ in AW2016 to 50 μg m$^{-3}$ in AW2017) than the grids coincident with the monitoring network sites. Regardless, both values exceed the 25% target set for Beijing (MEE, 2017).

Figure 8 further compares observed and modelled relative changes in PM$_{2.5}$. This confirms the skill of the model at reproducing the relative change in PM$_{2.5}$, despite an overall positive bias in total PM$_{2.5}$ (Figs. 4 and 7) and individual PM$_{2.5}$ components (Fig. 5). The modelled relative changes in PM$_{2.5}$ are somewhat more spatially consistent with the observations in BTH ($r = 0.82$) than the surrounding area ($r = 0.76$). The variance is closer to
unity for BTH (slope = 0.84) than the surrounding area (slope = 0.74). The relative change in PM$_{2.5}$ outside the emission control domain is -10$\%$ according to the observations, and -11$\%$ in the coincident model grids. During the emission control period, the area surrounding BTH in the domain shown in Fig. 7 is likely influenced by anthropogenic emissions from neighbouring regions that are the same in AW2017 and AW2016. This may account for the differences in the direction of change for 14 model grids (decline) compared to the observations (increase) in Fig. 8.

The emissions that we estimate for the control period (AW2017) are also shown in Fig. 6. According to our approach, total emissions reductions in AW2017 compared to AW2016 in BTH are 0.27 Tg NO$_x$, 0.66 Tg SO$_x$, 9.7 Tg CO, 70 Gg OC, and 50 Gg BC. Emissions of NH$_3$ and NMVOCs are unchanged due to limited constraints on these. There is an Ammonia Monitoring Network in China (AMoN-China), but there are only 8 sites in BTH, and the data are not publicly available (Pan et al., 2018). Relating NH$_3$ concentrations to emissions is also complicated by its ability to partition to sulfate aerosols (Fu et al., 2017) that decline by 36$\%$, according to the model, in AW2017 relative to AW2016 due to decline in SO$_2$ emissions. There were also no mandatory measures targeting NH$_3$ sources during AW2017. Agricultural activities such as fertilizer and livestock excreta (Huang et al., 2012), dominant NH$_3$ sources in China, are also at a minimum in autumn and winter (Kong et al., 2019). Zhang et al. (2021) reported that NH$_3$ emissions were relatively unchanged in almost half of the 28 cities and varied in others from a 35$\%$ decrease to a 33$\%$ increase. Controls in BTH in AW2017 should have targeted NMVOCs sources, but these have limited effect on PM$_{2.5}$ according to GEOS-Chem. The sensitivity simulation we conducted with a 50$\%$ increase in NMVOCs emissions only causes a 1$\%$ increase in surface concentrations of PM$_{2.5}$. Mechanisms leading to the formation of wintertime SOA are not well understood. Field campaigns in winter in Beijing suggest significant SOA formation from oxidation of NMVOCs (Li et al., 2020a; Li et al., 2020b), whereas Wang et al. (2021) propose that rapid aqueous-phase oxidation of primary OA dominates SOA formation and would resolve the observed decline in SOA in Beijing in winter at the same time that NMVOCs emissions have remained relatively constant.

We also quantify the contribution of meteorology to the decline in PM$_{2.5}$ in AW2017. To do this, we compare modelled PM$_{2.5}$ from the AW2017 simulation (Fig. 7) to a simulation with AW2017 emissions and AW2016 meteorology. The decline in PM$_{2.5}$ in BTH due to differences in AW2016 and AW2017 meteorology only is 12$\mu$g m$^{-3}$ or 57$\%$ of the decline in PM$_{2.5}$ from changes in both emissions and meteorology. The contribution of meteorology is similar, 54$\%$, in the surrounding area. Our estimate is slightly less than the city-scale study by Zhang et al. (2019). They attribute 70$\%$ of the decline in PM$_{2.5}$ in BTH to variation in meteorology using CMAQ driven with WRF meteorology and the national MEIC and regional Beijing emissions inventories. Zhang et al. (2021) identified large variability in the contribution of emissions and meteorology to decline in PM$_{2.5}$ across the 28 cities, of 2-82$\%$ for emissions and 18-98$\%$ for meteorology. We find that the meteorological factors contributing to decline in PM$_{2.5}$ in BTH in AW2017 include dilution and dispersion of pollution due to stronger south-easterly winds and a 7$\%$ higher planetary boundary layer in AW2017 than AW2016. Efficiency of formation of secondary inorganic aerosols would also be less in AW2017 than AW2016 in response to 5$\%$ lower RH.

Interannual variability in RH has similarly been identified as the cause for sustained air pollution over BTH despite a dramatic decline in precursor emissions from lockdown measures imposed to mitigate the spread of the SARS-
CoV-2 virus (Le et al., 2020). The meteorological factors that we identified with GEOS-Chem are consistent with those identified by Zhang et al. (2021) using WRF.

5 Conclusions

Strict emission controls were implemented across 28 cities in and around the Beijing-Tianjin-Hebei region (BTH) in autumn-winter 2017/2018 to address severe air pollution, specifically fine particles (PM$_{2.5}$). We used national and local surface monitoring network observations of PM$_{2.5}$ and trace gases with the GEOS-Chem model to assess the efficacy of emission controls, following evaluation of the network with independent measurements.

PM$_{2.5}$ and trace gases (NO$_x$, SO$_2$, and CO) from the surface networks are temporally consistent with independent measurements ($r > 0.9$ for PM$_{2.5}$ and $r > 0.7$ for gases) and exhibit discrepancies that are in large part due to variability of these in the urban environment. According to these networks, PM$_{2.5}$ in BTH decreased by 28% from 103 µg m$^{-3}$ to 75 µg m$^{-3}$ in the control period relative to the previous year, exceeding the regional target of 15%.

The model with emissions scaled to address large biases in NO$_x$, SO$_2$, and CO emissions, reproduces the spatial distribution in PM$_{2.5}$ ($r = 0.68$). Despite a 15% positive bias in total PM$_{2.5}$ and large biases in PM$_{2.5}$ composition, the model captures the relative decline in PM$_{2.5}$ in BTH of 25%. According to the model constrained with the network measurements, decline in emissions in BTH due to strict controls are 0.27 Tg NO$_x$ as NO, 0.66 Tg SO$_2$, 9.7 Tg CO, 70 Gg OC, and 50 Gg BC. These account for less than half the observed decline in PM$_{2.5}$ and alone lead to an 8% reduction in PM$_{2.5}$, falling shorting of PM$_{2.5}$ reduction target. The remainder is due to meteorology, specifically a deeper planetary boundary layer, stronger winds, and lower relative humidity during the control period.

This supports the need for much stricter emissions controls in BTH and other parts of China where these controls are now adopted.

Data Availability

The GEOS-Chem model outputs used in this study are available at:

https://github.com/GongdaLu/BTH_emission_control.

Author Contributions

GLu performed the GEOS-Chem simulations, analysed the model and measurement data, and prepared the manuscript. EAM assisted in the writing and provided supervisory guidance, with co-supervision and editorial contributions from ZS. JDL provided APHH gas concentration data. ZS, TVV and JX performed PM composition analyses and provided guidance on using APHH data. QZ provided the original MEIC emission inventory that LS processed for input to GEOS-Chem. GLuo and FY provided source codes for the updated wet scavenging scheme in GEOS-Chem.

Competing Interests

The authors declare that they have no conflicts of interest.
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References


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Figure 1. Evaluation of local and national network PM$_{2.5}$ measurements. Points are hourly PM$_{2.5}$ measurements. The top row compares CNEMN to the APHH urban site (left), and CNEMN to the US Embassy in autumn-winter 2016/2017 (AW2016) (centre) and 2017/2018 (AW2017) (right). The bottom row compares BJMEMN to the APHH urban site (left), and BJMEMN to the US Embassy in AW2016 (centre) and AW2017 (right). Reduced major axis (RMA) regression statistics, Pearson’s correlation coefficients (r), and the percent difference (Diff = monitoring network minus independent measurement) are given. Lines are the RMA regression (red) and 1:1 line (blue dashed).
Figure 2. Evaluation of local and national network trace gas concentration measurements. Points are hourly measurements. Panels compare CNEMN (top) and BJMEMN (bottom) NO$_2$ (left), SO$_2$ (centre) and CO (right) to the APHH urban site in November-December 2016. RMA regression statistics, Pearson’s correlation coefficients ($r$), and the percent difference (Diff = monitoring network minus independent measurement) are given. Lines are the RMA regression (red) and 1:1 line (blue dashed). The green dashed line is the reported monitoring network instrument detection limit (MEE, 2012).
Figure 3. Change in surface air pollution in BTH and the surrounding area in AW2017 relative to AW2016. Individual points are monitoring network site changes for the target region (triangles within area shaded grey) and the surrounding area (circles in the non-shaded area). Values inset give the percent change for sites in the grey domain only. Note the colourbar is uneven.
Figure 4. Evaluation of GEOS-Chem simulation of air pollutant concentrations in AW2016. Observations are averaged onto the GEOS-Chem grid. Points are simulated and observed NO\textsubscript{2} (top left), SO\textsubscript{2} (top right), CO (bottom left) and PM\textsubscript{2.5} (bottom right) in the entire domain in Figure 3 for grid squares with at least three coincident surface sites. Points are coloured by the number of surface sites. The model uses scaled MEIC emissions (see text for details). RMA regression statistics, Pearson’s correlation coefficients (r), and model normalized mean biases (NMB) are given. Lines are the RMA regression (black) and 1:1 line (blue dashed).
Figure 5. Relative contribution of individual PM$_{2.5}$ components during the APHH campaign. Panels are measured (left) and modelled (right) percent contribution for the urban IAP (top) and rural Pinggu (bottom) sites. Components are sulfate, nitrate, ammonium, OC, BC and Other. Other is the sum of trace metals, mineral dust and other ions, the non-carbon portion of OC, and aerosol water at 33–35% relative humidity (RH) for the measurements and 35% RH for the model. Values above the pies are mean total PM$_{2.5}$. 

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Figure 6. Total anthropogenic emissions in BTH in AW2016 and AW2017. Emissions are for grids covering the grey shaded area in Figure 3. Emissions are from the MEIC with scaling factors to address discrepancies between the model and observations for AW2016 and to reproduce the change in air pollutant concentrations in the AW2017 emission control period (see text for details). Vertical axes are emissions for NO, BC, OC, and SO$_2$ on the left axis and for CO on the right axis.

Figure 7. Spatial distribution of absolute and relative changes in observed and modelled PM$_{2.5}$ before and during the strict emission control period. The observed (shapes) and modelled (background) PM$_{2.5}$ are shown for AW2016 (left) and AW2017 (centre). The right panel is the relative change in observed and modelled PM$_{2.5}$. Observations are distinguished as within (triangles) and outside (circles) the emission control region (grey area in Figure 3). Inset values are the observed (OBS) and modelled (GC) means for sites and coincident grids in the emission control domain. Note the uneven colour scale in the right panel.
Figure 8. Comparison of observed and modelled relative changes in PM$_{2.5}$. Triangles indicate data in BTH (grey area in Figure 7) and circles indicate data outside BTH. These are coloured by the number of sites in each GEOS-Chem grid. The RMA regression statistics and lines, Pearson’s correlation coefficients (r), and NMB are given for all points (black text) and for those in BTH (green text).