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Factors affecting recent $PM_{2.5}$ concentrations in China and South Korea from 2016 to 2020

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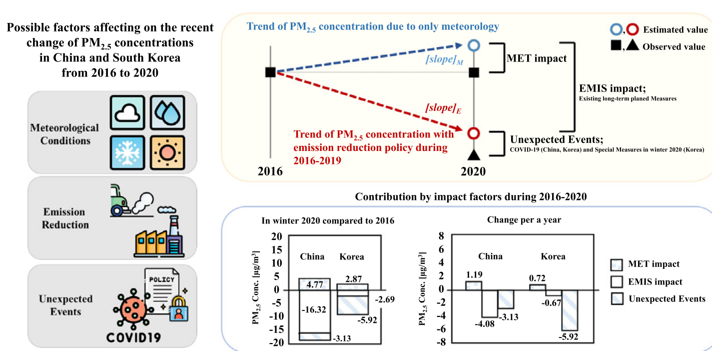
HIGHLIGHTS

- The contribution of possible factors to the recent change of PM2.5 concentrations in China and South Korea is investigated quantitatively
- Meteorological conditions, emission reduction polices, and unexpected events are considered as possible major factors.
- The effects of meteorological conditions have increased $PM_{2.5}$ concentrations during winter 2016-2020 in China and South Korea.
- The existing and long-term planed emission control policies are effectively implemented to reduce the elevated $PM_{2.5}$ concentration levels
- The unexpected events such as the COVID-19 pandemic also have the significant impact on the decrease in $PM_{2.5}$ concentrations in winter 2020

GRAPHICAL ABSTRACT



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ABSTRACT

This study used observational data and a chemical transport model to investigate the contributions of several factors to the recent change in air quality in China and South Korea from 2016 to 2020. We focused on observational data analysis, which could reflect the annual trend of emission reduction and adjust existing emission amounts to apply it into a chemical transport model. The observation data showed that the particulate matter ($PM_{2.5}$) concentrations during winter 2020 decreased by −23.4 % (−14.68 μg/m^3) and − 19.5 % (−5.73 μg/m^3) in China and South Korea respectively, compared with that during winter 2016. Meteorological changes, the existing national plan for a long-term emission reduction target, and unexpected events (i.e., Coronavirus disease 2019 (COVID-19) in China and South Korea and the newly introduced special winter countermeasures in South Korea from 2020) are considered major factors that may affect the recent change in air quality. The impact of different meteorological conditions on $PM_{2.5}$ concentrations was assessed by conducting model simulations by fixing the emission amounts; the results indicated

Abbreviations: ASOS, Automated Synoptic Observing System; CMAQ, Community Multiscale Air Quality; COVID-19, Coronavirus disease 2019; CTM, chemical transport model; GTS, Global Telecommunication System; JFM, January, February, and March; MCIP, Meteorology-Chemistry Interface Processor; MEGAN, Model of Emission of Gases and Aerosols from Nature; NCEP FNL, National Center for Environmental Prediction-Final data; NIER, National Institute of Environmental Research; OBBC, Observation Based Bias Correction; T2, temperature; WRF, Weather Research and Forecasting; WS, wind speed.

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changes of +7.6 % (+4.77 μg/m^3) and + 9.7 % (+2.87 μg/m^3) in China and South Korea, respectively, during winter 2020 compared to that during winter 2016. Due to the existing and pre-defined long-term emission control policies implemented in both countries, $PM_{2.5}$ concentration significantly decreased from winter 2016–2020 in China (−26.0 %; −16.32 μg/m^3) and South Korea (−9.1 %; −2.69 μg/m^3). The unexpected COVID-19 outbreak caused the $PM_{2.5}$ concentrations in China to decrease during winter 2020 by another −5.0 % (−3.13 μg/m^3). In South Korea, the winter season special reduction policy, which was introduced and implemented in winter 2020, and the COVID-19 pandemic may have contributed to −19.5 % (−5.92 μg/m^3) decrease in $PM_{2.5}$ concentrations.

1. Introduction

China and South Korea have suffered from severe air pollution, resulting in environmental, social, and political problems and adversely affecting human health owing to exposure to high concentrations of air pollutants (Choi et al., 2019; Matus et al., 2012; Hwang et al., 2020; Song et al., 2017). Therefore, contingency responses are being implemented through various national policies to improve air quality in both countries.

According to a previous study analyzing long-term trends in $PM_{2.5}$ concentrations, the average concentrations of $PM_{2.5}$ in China had increased by 0.04 μg/m^3/yr during the period of 2001–2005, while it started to decrease by −0.65 μg/m^3/yr and − 2.33 μg/m^3/yr during 2006–2010 and 2011–2015, respectively (Lin et al., 2018). Looking closely into the decreasing trend from 2013 to 2017, national emissions of SO_2, NO_X, and $PM_{2.5}$ decreased by −59 %, −21 %, and − 33 %, respectively, through the 2013–2017 Air Pollution Prevention and Control Action Plan implemented in China (Zhang et al., 2019). The $PM_{2.5}$ concentration decreased by −6.9 % in 2018 compared to that in 2017 and by −18.2 % in 2019 compared to that in 2018, owing to the effect of the Action Plan for Comprehensive Treatment of Air Pollution in Autumn and Winter in the Jing-Jin-Ji region and the surrounding areas (Liu et al., 2022).

Examining the long-term trend of $PM_{2.5}$ concentrations in South Korea, a decreasing trend has been observed over 15 years (2003–2017), while temporal increases of $PM_{2.5}$ concentrations (2003–2007 and 2012–2016) in Seoul area have weakened its long-term declining trend (Kim et al., 2020). As in China, South Korea has also been making efforts to improve air quality with the national target to keep annual $PM_{2.5}$ concentration below 16 μg/m^3 in 2024, a − 35 % decrease as compared to 26 μg/m^3 in 2016 (MOE, 2019). Especially, during winter when high concentration events occur frequently over East Asia, the special reduction countermeasure has been implemented from December 2020 to March 2021; as a result, the frequency of severe high concentration events is significantly reduced (MOE, 2021).

Meteorological conditions also play a key role in transport and diffusion processes and atmospheric chemical reactions. In particular, a high pressure system in winter stabilizes the atmosphere and provides more favorable conditions for the accumulation of emitted air pollutants, extensively elevating their air concentrations in Northeast Asia as compared to that in other seasons (Lee et al., 2013; Oh et al., 2015). This is one of the possible reasons why $PM_{2.5}$ high concentration events occur more frequently during winter in China and South Korea, besides the addition of elevated emissions induced by the increased demand of local heating (Zhang et al., 2022b; Itahashi et al., 2017; Yin et al., 2019b). Considering that the westerly winds are the prevailing synoptic winds during winter over Northeast Asia, it is evident that the long-range transport could significantly contribute to severe high concentrations of $PM_{2.5}$ over the Korean peninsula located in the downwind area (Oh et al., 2015; He et al., 2003; Kotamarthi and Carmichael, 1990). Moreover, the change of wind speed and direction, humidity, and the planetary boundary layer height are key factors affecting the regional air quality (Li et al., 2019; Tian et al., 2014; Yin et al., 2019a).

It should be noted that unexpected events have affected air quality in both China and South Korea in 2020. An outbreak of the Coronavirus disease 2019 (COVID-19) pandemic was identified in China at the end of 2019 (Zhou et al., 2020; Wu et al., 2020), and the pandemic remains active worldwide. It has led to vast changes in society, the economy, and the environment (Hiscott et al., 2020; Fernandes, 2020; Helm, 2020). Consequently, the unprecedented decrease of anthropogenic emission and atmospheric concentration of air pollutants in 2020 has been reported, as the pandemic triggered reduced social and economic activities as a result of travel restrictions, lockdowns, business restrictions and closures, etc. (Lian et al., 2020; Ropkins and Tate, 2021; Zangari et al., 2020; Ju et al., 2021; Kroll et al., 2020).

The national policies to improve the air quality, the temporary and climatological change of meteorology, and the unexpected social/economic events all resulted in extensive changes in the emission of air pollutants and a different sensitivity on the atmospheric concentration of air pollutants following combined or separate changes (Sofia et al., 2020; Wang et al., 2010; Zheng et al., 2015).

We attempted to quantitatively assess the factors that could contribute to the total changes in $PM_{2.5}$ concentrations in China and South Korea during winter 2020 compared to that during winter 2016. Observational data were used to reflect the annual/monthly trend of emission reduction and adjust existing emissions to incorporate them into a chemical transport model. To prepare emission data, the bottom-up approach, which is one of the main inputs for a chemical transport model, has a limitation in the practical sense; it is too slow to reflect the actual amount of emission in time because it requires the update of several social and economic statistics describing human activities in detail. Therefore, previous studies introduced the adjusted emission data by using the temporal change of observed surface concentration (Lamsal et al., 2011; Huang et al., 2015). This method can be advantageous in reducing the uncertainty of a chemical transport model with sufficient consideration for the rapidly changed emission tendency (Napelenok et al., 2011; Holnicki and Nahorski, 2015; Zheng et al., 2009). Previous studies (Bae et al., 2021; Cheng et al., 2019; Seo et al., 2018) also pointed out that it might be vital to make a quantified assessment of the impact of reduction policies and other factors influencing a change in air quality status and to provide a reliable scientific basis for decision-makers by using the results of a chemical transport model. With this type of scientific motivation and policy-relevant purpose, we conducted a chemical transport model based on scenario-based simulations with each differently adjusted emission dataset to separate each different factor that affects the change of the total concentrations.

2. Material and methods

2.1. Surface observation data

Meteorological data of China and South Korea were collected from the Global Telecommunication System (GTS) and Automated Synoptic Observing System (ASOS) to evaluate the performance of the meteorological model. Surface mass concentration data measured at the ambient monitoring sites were used to analyze the current temporal trend and spatial distribution of $PM_{2.5}$ and to evaluate the performance of air quality simulations. Observed surface mass concentrations of air pollutants were also applied to adjust the pre-defined emission inventory. For observations in China, we collected the data for the hourly concentrations of $PM_{2.5}$, PM_{10}, NO_2, SO_2, CO, and O_3 at 1436 monitoring stations through the China National Urban Air Quality Real-time Publishing Platform. Considering Korean data, we obtained the same type of observations as for China at 505 monitoring stations on the Urban Monitoring Network operated by the National Institute of Environmental Research (NIER), Korea. The locations of air quality monitoring stations over China and South Korea are shown in Fig. 1.

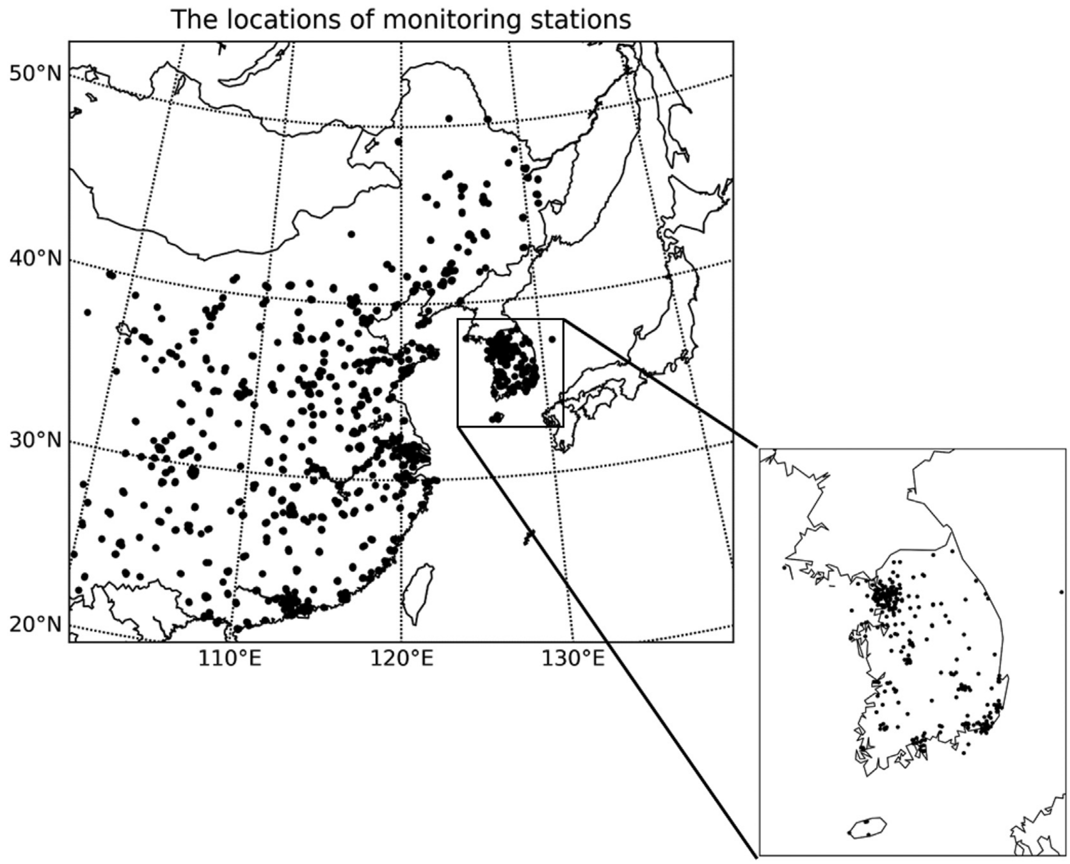

Fig. 1. Modeling domain (horizontal resolution of 27 km × 27 km and 174 × 128 cells). Black dots indicate air quality monitoring stations over China and South Korea.

2.2. Emission adjustment

There are technical difficulties in updating the appropriate emission data annually because the required information for estimating the emission amount is unavailable in real-time or nearly real-time from the countries and cities. This is evidently considered as one of main causes underlying the increased uncertainty of the simulation results of the chemical transport model. Therefore, we made an observation-based adjustment of the pre-defined emission amounts that were estimated using the socio-economic statistics of the reference year 2016, to resolve the temporal trend and spatial distribution change of emission amounts during the target period of January, February, and March from 2017 to 2020. The reference year has been selected as 2016 for applying the atmospheric chemistry transport model, as it is the most recent year for which updated emission data may be available. The year of 2016 holds particular significance for air quality research in South Korea, as it marks the KORUS-AQ campaign – an intensive field campaign designed to investigate the emission sources and chemical mechanism affecting air quality in the Seoul Metropolitan Area (Crawford et al., 2021; Schroeder et al., 2020; Choi et al., 2019). The KORUS-AQ campaign used a combination of ground-based, aircraft, and satellite measurements to collect data on various air pollutants, including nitrogen oxides (NOx), various volatile organic compounds (VOCs), sulfur dioxide (SO_2), and chemical composition of fine particulate matter ($PM_{2.5}$). Especially, a variety of aircraft, including NASA's DC-8 and King Air B-200, as well as the Korean B-200, were equipped with a suite of instruments to measure trace gases, aerosols, and cloud properties and deployed to collect air quality data during the campaign. It also included ground-based measurements using in-situ and remote sensing instruments. These ground-based measurements provided complementary data to the airborne measurements, allowing researchers to gain a more comprehensive understanding of the air pollution situation in South Korea.

To apply the method for adjusting emissions data using observational data (Feng et al., 2022; Bae et al., 2022), we used KORUS version 5 emission inventory developed by Konkuk University (Simpson et al., 2020; Oak et al., 2019) as the pre-defined emission in the reference year, 2016.

The key of the adjustment method is to establish the rate of emission change during the target period compared to that during the reference year. The first step involves separating the meteorological contribution from all the current changes in air concentrations. We calculated the meteorological impact on the change of total mass concentrations by using a chemical transport model simulation with a fixed emission but temporally different meteorological conditions. As the emission data were fixed in 2016, the meteorological impacts (MET_{yyyy}) for each year of the target period could be estimated by calculating the normalized variation of a chemical transport model simulation result (MOD_{E2016}^{Myyyy}) with each different meteorological condition in a specific target year (yyyy) compared to one in 2016 (MOD_{E2016}^{M2016}). In this step, in order to remove chemical nonlinear impacts of meteorology on difference chemical species much as possible and consider only diffusion and transport effects, we fixed the same emissions for CO, which is commonly known as a trace gas due to its long chemical lifetime and quantified the meteorological effects by varying the meteorological conditions only instead.

$$MET_{yyyy} = \frac{MOD_{E2016}^{Myyyy} - MOD_{E2016}^{M2016}}{MOD_{E2016}^{M2016}} \times 100 \tag{1}$$

The next step involves projecting the expected concentrations in a target year (yyyy) by adding the incremental change of the observed concentration (OBS_{2016}) in 2016 due to the meteorological impact (MET_{yyyy}) for each year of the target period.

$$\text{Expected Conc.}_{yyyy} = OBS_{2016} + (OBS_{2016} \times MET_{yyyy}) \quad (2)$$

If we consider that the expected concentration obtained above could resolve only the meteorological impact, it would be quite straightforward to separate the impact of emission changes by subtracting the expected concentration from actual observation of concentration (OBS_{yyyy}), which includes both the meteorological and the emission change. Finally, we can obtain the rate of emission change ($EMIS_{yyyy}$) for each target year compared to 2016 by subtracting the expected from the actual observations.

$$EMIS_{yyyy} = \frac{OBS_{yyyy} - \text{Expected Conc.}_{yyyy}}{OBS_{2016}} \times 100 \quad (3)$$

2.3. Model configurations and input data

To assess the several factors that have impacted the current change in air quality in China and South Korea, a chemical transport model (CTM) was applied. The target seasons of CTM simulations in this study were the winters (January, February, and March) from 2016 to 2020. Model simulations were conducted using two distinct types of emission datasets (KORUSv5 and Adjusted KORUSv5) for each year. To provide meteorological data as an input to CTM, the Weather Research and Forecasting (WRF) model (Skamarock et al., 2005) version 3.9.1.1 and WPS version 4.1 were used. WRF modeling was performed for a month with 3 days of spin-up, and the National Center for Environmental Prediction-Final data (NCEP FNL; NCEP, 2000) with $1^\circ \times 1^\circ$ of the spatial resolution was used as the initial and boundary conditions, respectively, for WRF simulations. WRF outputs were post-processed to be available in the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006) version 5.2 through the Meteorology-Chemistry Interface Processor (MCIP; Otte and Pleim, 2010) version 4.4. For anthropogenic emissions, KORUSv5, which represents the emission amounts of the reference year 2016, was adjusted in each target year by using the method described in section 2.2, and the biogenic emissions were also estimated with the Model of Emission of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006) version 2.1. The Carbon Bond version 6 (CB6r3; Wyat Appel et al., 2016) was adopted as the gas phase chemical mechanism and the AERO6 was adopted for the particulate matter. The model domain for WRF and CMAQ included the northeastern part of China and the entire Korean peninsula, with a horizontal grid size of 27 km (Fig. 1). Detailed configurations of the WRF and CMAQ models are shown in Table S1.

2.4. Consideration of possible impact factors on air quality

To consider the possible impacts on the current air quality in China and South Korea, we attempted to separate each influence of emission, meteorology, and an unexpected event by using observational data and WRF/CMAQ simulation results with two different emissions as shown below.

(1) $EMIS_{Fixed}$: Simulations were performed with fixed emission input and variable meteorological conditions for each target period by month and the first three days were used as spin-up times.
(2) $EMIS_{Adj}$: Same as $EMIS_{Fixed}$; however, the emission input was not fixed and changed according to the target period. Note that the adjusted emission was calculated using the Observation Based Bias Correction (OBBC) method described in section 2.2.

Using the change in $PM_{2.5}$ concentrations in the observation data and model results from 2016 to 2020, we were able to separate the contributions of different impact factors to the current air quality change in China and South Korea. The observational variation of $PM_{2.5}$ concentrations during the winter seasons from 2016 to 2020 can reflect the total changes combined with all the impacts of both different meteorological conditions and emission reductions. The change in $PM_{2.5}$ concentrations from the model simulation with $EMIS_{Fixed}$ in each year is supposed to be only due to a different meteorological condition in each year compared with that in 2016. The impact of emission reduction, including unexpected events, can also be assessed by calculating the difference between model simulations and applying the $EMIS_{Fixed}$ and $EMIS_{Adj}$ in each target year.

3. Results

3.1. Observational change of $PM_{2.5}$ concentrations from 2016 to 2020

The mean $PM_{2.5}$ concentrations observed in China and South Korea during the annual and winter seasons (JFM; January, February, and March) from 2016 to 2020 are shown in Fig. 2.

In China, the annual mean $PM_{2.5}$ concentrations have sharply decreased by −23.7 % from 48.80 μg/m^3 in 2016 to 37.22 μg/m^3 in 2020. The $PM_{2.5}$ concentrations during winter (JFM) were almost 11–14 μg/m^3 higher than the annual mean and decreased by −23.4 % from 62.79 μg/m^3 to 48.11 μg/m^3 during 2016–2020. Considering the annual mean $PM_{2.5}$ concentrations, South Korea also experienced the decreasing trend over the past 5 years, from 29.45 μg/m^3 in winter 2016 to 23.72 μg/m^3 in winter 2020, although the rate of decrease (−19.5 %) is much lower than that in China. It is noteworthy that the concentrations in JFM of each year compared to the previous year have repeatedly increased or decreased rather than continuously decreased for 5 years: +2.47 μg/m^3 (+8.4 %), −1.01 μg/m^3 (−3.2 %), +4.82 μg/m^3 (+15.6 %), −12.01 μg/m^3 (−33.6 %) for 2017–2020, respectively. Previous studies (Chen et al., 2015; Kim et al., 2017) already mentioned that the concentrations during winter are shown to be much higher than the annual concentrations in the northeast Asian region. It is well known that the very high concentration events usually occur during winter over the northeast Asian region, indicating that the reduction policy for the high concentration has only been effective to avoid the increasing trend of the concentrations, but not to establish a decreasing trend. It can also be observed that the sharply decreased concentrations in 2020 occurred during the winter season, possibly because of the COVID-19 pandemic in China and South Korea.

3.2. Reduction rate of emission adjustment

By applying the adjustment process described in Section 2.2, we show the estimated rate of emission change of NO_2, SO_2, and CO for each target month/year compared with that in 2016 in China and South Korea in Table 1. The reduction rate of emission amounts for each species calculated in this study seems to be quite comparable to that from previous studies. For example, Zheng et al. (2018) reported that the emission amounts of NO_2 and SO_2 in China had been reduced by approximately −2 % and − 11 % annually in 2017 compared with that in 2016, respectively; whereas, in our study, the emissions changed from −9 % to +15 % (NO_2) and − 11 % to −23 % (SO_2) for January to March 2017, respectively. Considering a long-term change, it is also reported that SO_2 decreased by approximately −56 % to −78 %, and NO_2 decreased by −22 % to −51 % in 2020 compared with that in 2016 (Wang et al., 2022), respectively; whereas this study shows a − 58 % to −86 % and − 27 % to −48 % reduction in SO_2 and NO_2 from January to March 2020, respectively.

According to the bottom-up emission amounts (2016–2019) announced by the Ministry of Environment, Korea (https://www.air.go.kr/capss/emission), the reductions in 2019 compared with that in 2016 have been −24 %, −13 %, and − 5 % for SO_X, NO_X, and CO, respectively. This data is similar to the adjusted amounts estimated in this study: −27 % to −32 % (SO_2), −7 % to −17 % (NO_2), and − 3 % to −11 % (CO). It is noteworthy that the bottom-up emission represents an annually averaged value, but this study calculates the data only for winter months (January to March) of 2016–2020.

In addition, we conducted an evaluation of the adjusted emission using satellite observation data, tropospheric NO_2 column density from the

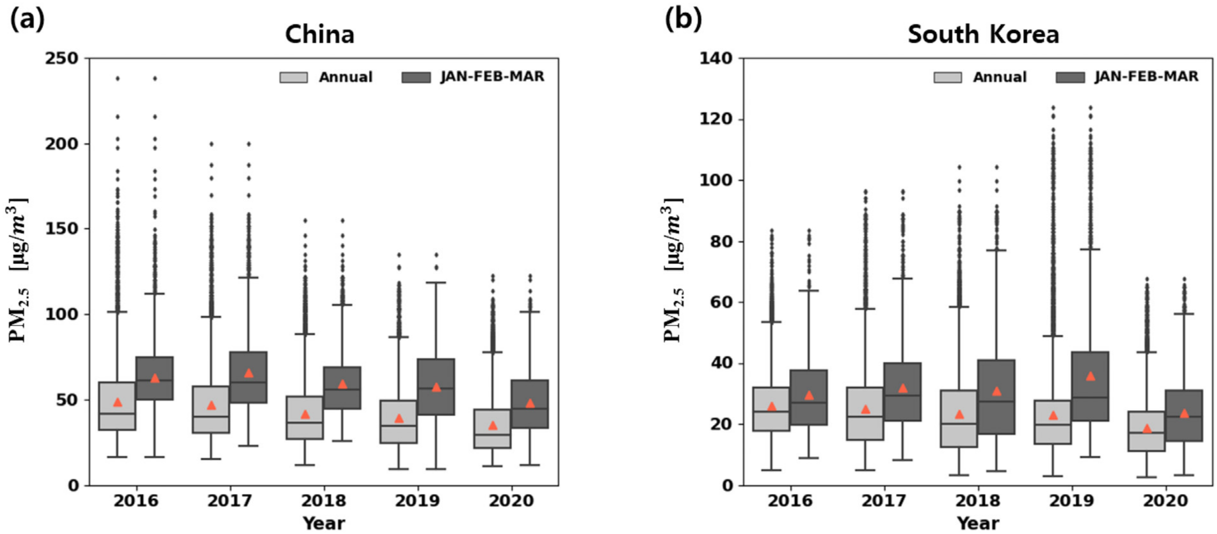

Fig. 2. Boxplot of $PM_{2.5}$ concentration changes in China (a) and South Korea (b) from 2016 to 2020 using observational data. Light-gray and dark gray represents the annual average and the average of January, February, and March, respectively. Red triangles indicate the average points.

Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite (Fig. S1, Table S2). It has been shown that the average NO_2 column density during January, February, and March of 2020 compared to the same period in 2016 has decreased by approximately −33 % in China and − 13 % in South Korea. Considering that synoptic weather patterns and boundary layer heights during the two periods were similar (Fig. S2), it should be noted that the reduction rate shown in the satellite data is comparable to the reduction rate of the adjusted emissions in our study, which was approximately −27 % to −48 % in China and − 15 % to −19 % in South Korea.

KORUSv5, the emission data used as the 2016 reference emissions before adjustment in this study, is the latest and scientifically best available source-specific emission data, and has been developed and updated using various observational data and model validation results from the KORUS-AQ field campaign, 2016 conducted to better understand air pollution in Korea and East Asia (Fried et al., 2020; Kwon et al., 2021; Park et al., 2021).

In particular, VOCs still need to be more rigorously developed and validated through follow-up studies, but significant improvements (KORUSv1➔KORUSv5) have been made especially for aromatic species (i.e., Benzene, Toluene, Ethylbenzene, Xylene and Isoprene) emissions by incorporating airborne and satellite observations during the campaign. This study used KORUSv5 emissions which reflect the activities of all available sources including paints, solvents, cooking, etc., as the 2016 baseline emissions of VOCs, while the recent temporal rates of change were just estimated using observations of CO concentrations. The reason for adjusting the 2020 emissions of VOCs using the temporal changes in CO concentrations in this study is that it has a relatively long chemical lifetime and its chemical oxidation in the lower atmosphere is known to be very slow (Jaffe, 1968), so as to be known to represent socio-economic anthropogenic activity as accurately as possible.

Table 1
Results of emissions adjustment calculations based on the Observation Based Bias Correction method. The adjustment amount of NO_2, SO_2, and CO was calculated by year (2017–2020), month (Jan, Feb, Mar), and country (China, South Korea).

Year	Month	China			South Korea		
		NO_2 (%)	SO_2 (%)	CO (%)	NO_2 (%)	SO_2 (%)	CO (%)
2017	Jan	−9.320	−23.280	−8.671	−2.309	−12.459	−1.689
	Feb	15.532	−11.145	−3.188	7.516	−2.107	5.397
	Mar	−2.739	−23.245	−13.581	−1.809	−1.761	4.394
2018	Jan	−1.487	−44.673	−21.514	−4.792	−20.326	−6.783
	Feb	−1.761	−39.134	−17.243	0.218	−11.200	−3.054
	Mar	−4.407	−39.616	−13.321	−8.465	−11.948	1.329
2019	Jan	−12.323	−68.074	−30.620	−6.939	−31.927	−10.698
	Feb	−20.590	−65.819	−24.848	−5.882	−32.419	−5.994
	Mar	−13.744	−56.988	−29.655	−17.099	−26.745	−3.161
2020	Jan	−39.669	−86.024	−43.288	−15.398	−45.455	−11.749
	Feb	−47.555	−75.087	−41.822	−19.058	−51.014	−17.796
	Mar	−26.933	−58.584	−32.778	−16.404	−25.665	−2.539

3.3. Evaluation of model performance

Various statistical metrics were used to evaluate the model simulation performance in this study. Table S3 summarized the comparison metric for temperature at 2 m (T2) above the ground and wind speed at the 10 m level (W10) between WRF model outputs and observed data at Beijing and Seoul. The WRF model seems to sufficiently reproduce the observed T2 and WS with mean biases within ±0.8 °C and ± 0.2 m/s, respectively, and the temporal correlation coefficient ranged from 0.82 to 0.99 for Beijing and Seoul, respectively. The performance of the chemical transport model, CMAQ, is shown in Table S4 by comparing the simulated concentrations of $PM_{2.5}$, PM_{10}, NO_2, SO_2, CO, and O_3 to the measured values at the Chinese and Korean sites. The simulated concentrations (MOD_{E2016}^{M2016}) in the 2016 reference year tended to underestimate both in China and South Korea. For the 2016 KORUS-AQ period, various previous modeling studies have reported that uncertainty in emissions is a major cause of model underestimation. Goldberg et al. (2019) argued for the underestimation of KORUSv5 emissions compared to top-down emissions from satellite observations during the 2016 KORUS-AQ. In addition, Park et al. (2021) conducted an ensemble modeling study using eight different chemical transport models with the same KORUSv5 emissions and showed that the results underestimated $PM_{2.5}$ and O_3 in Korea by −22 % and − 7 %, respectively. Similarly, Bae et al. (2021) found that CMAQ model results using KORUSv5 emissions underestimated $PM_{2.5}$, SO_2, and NO_2 in South Korea by −24.9 %, −30.6 %, and − 17.3 %, respectively. The results of the CMAQ model in this study also show that it underestimates $PM_{2.5}$, SO_2, NO_2, and O_3 by −14 %, −5.6 %, −23 %, and − 3.5 %, respectively, which seem to be within the validation performance range of the results simulated using the same KORUSv5 emissions in previous studies aforementioned. Moreover, it is of scientific significance and policy relevance that this study just focused on the annual variation of $PM_{2.5}$ average concentrations during the winter season (January, February, and March), and the CMAQ model simulated the annual variation with very low bias (Fig. 3). We believe that the high level of agreement in the annual change

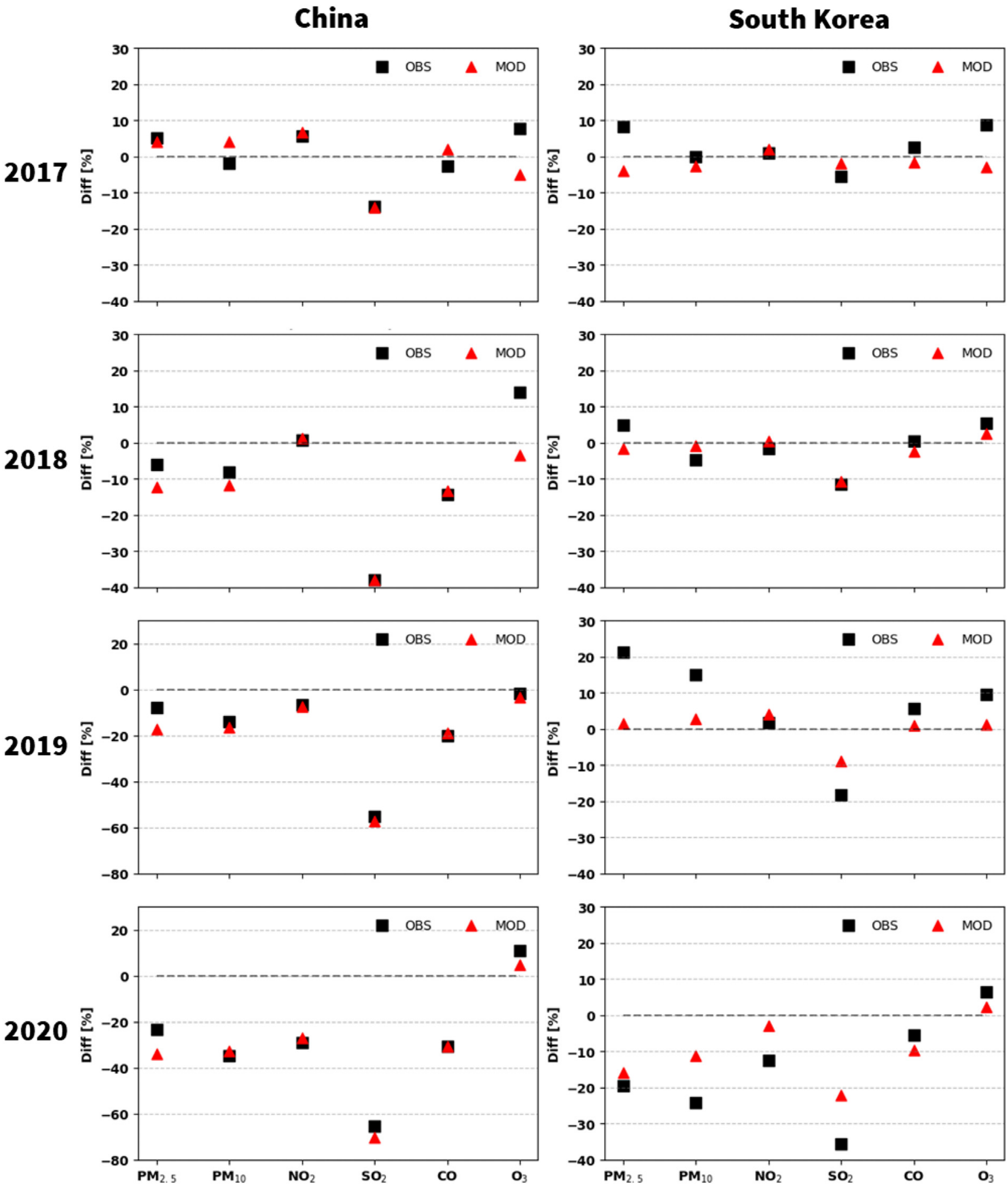

Fig. 3. $PM_{2.5}$ changes in the average concentration of Jan, Feb, and Mar for each year compared with that in 2016 (left column and right column indicate China and South Korea, respectively). Black squares indicate a change in the observed value and red triangles indicate a change in the model value.

itself of winter mean concentrations between the model and observational data allows for a reliability of the impact of the recent five-year changes in emissions and meteorology that we tried to investigate in this study.

In addition, we further evaluated the performance of the model results using measurement data of various chemical components of $PM_{2.5}$ (i.e., nitrate, sulfate, ammonium, organic carbon, and elemental carbon) at an intensive observation site located in Seoul during KORUS-AQ (Table S6). The results showed that sulfate and organic carbon were underestimated, while nitrate, elemental carbon, and ammonium were overestimated, with nitrate being the largest. Offsetting errors in aerosol modeling are a well-known feature of chemical transport models for a variety of complex reasons (Song et al., 2021; Wang et al., 2021). For example, it is known that NH_3 emissions are poorly estimated, and in regions where NH_3 is limited, models have so insufficient mechanisms as to consequently underestimate sulfate and overestimate nitrate (Chang et al., 2018; Fu et al., 2016). The limitations of chemical transport models are recognized as areas for improvement in future research but are beyond the scope of this study. For this reason, our investigation has focused on the incremental changes by reduction policies over the past five years in term of the total mass concentrations, rather than evaluating individual chemical components. However, it is crucial to emphasize that the results of this study should be carefully interpreted in conjunction with other relevant information and expertise perspective as for providing an optimal guidance for policy making on air quality management.

3.4. Impact of meteorological conditions

The impact of different meteorological conditions on the $PM_{2.5}$ concentrations in each year compared with that in 2016 was estimated by calculating the difference between MOD^{M2016}_{E2016} and MOD^{Myyyy}_{E2016} by fixing the emission to 2016 and altering the meteorology. Fig. 4 shows the year-to-year change in $PM_{2.5}$ concentrations (the zero-point corresponding to the $PM_{2.5}$ concentrations in 2016 as the base year) for three sensitivity cases; modeled values with fixed emissions (EMIS Fixed MOD), observed values (OBS), and observed values after removing meteorological effects (OBS–MET). The results of the model with fixed emissions show that $PM_{2.5}$ concentrations in both China and South Korea seem to increase and decrease, eventually rising in 2020 ($PM_{2.5}$ concentrations >0 in 2020). As for the meteorological impact of 2017–2020 compared to 2016, we found that meteorological conditions in 2017–2020 were favorable for increasing $PM_{2.5}$ concentrations, with an exception (−1.57 %, South Korea in 2017). In 2019, its maximum impact was observed in China (+13.26 %) and South Korea (+19.80 %) (Table 2(a)). For reflecting the meteorological impacts for each year shown in Table 2(a), the reconstructed concentrations (OBS–MET), which are the observed $PM_{2.5}$ concentrations minus the meteorological impacts, were calculated and displayed in Fig. 4. The observed (OBS) results show a continuous decreasing trend in $PM_{2.5}$ concentrations in China, while over South Korea, concentrations fluctuated and then decreased sharply in 2020. When meteorological effects were removed, the results (OBS–MET) showed a more significant decrease in China. In South Korea, $PM_{2.5}$ concentrations decreased significantly by 2020 and showed a slight downward trend over the five years, in contrast to the observations. The decreasing trends in $PM_{2.5}$ concentrations in both countries derived

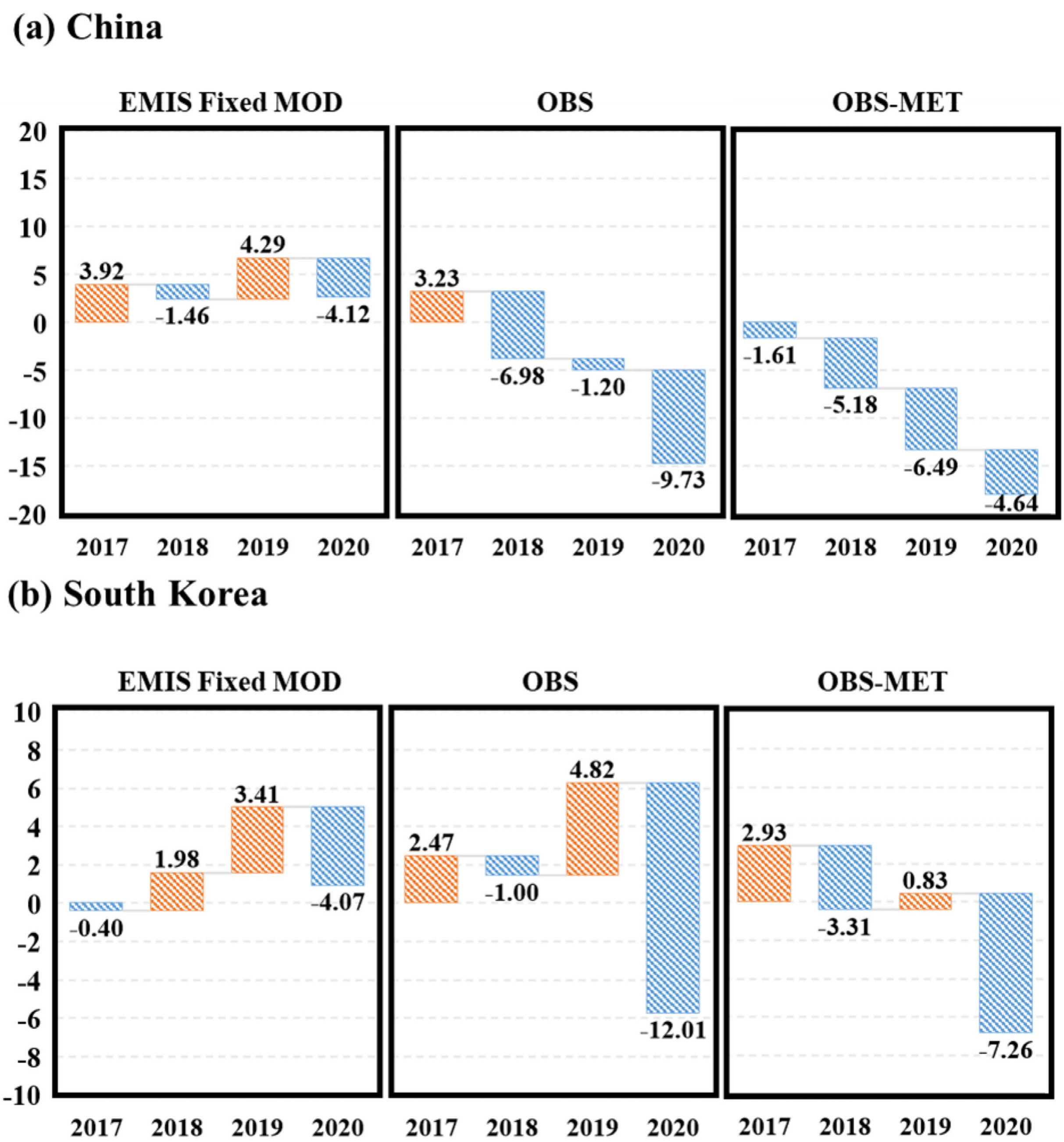

Fig. 4. Changes in average $PM_{2.5}$ concentrations in JAN, FEB, and MAR for year by year. Each bar and their values indicate the concentration change of $PM_{2.5}$ (μg/m^3) compared with previous year. The blue represents the decrease; the orange represents the increase. Also, a zero point indicates a value corresponding to 2016.

Table 2
Meteorological (a) and emission impacts (b) from 2017 to 2020 compared with that in 2016 in China and South Korea. Positive and negative values indicate the increasing and decreasing $PM_{2.5}$ concentrations, respectively.

		2017	2018	2019	2020
(a) MET	China	7.70	4.84	13.26	5.17
	South Korea	−1.57	6.28	19.80	3.68
(b) EMIS	China	−3.49	−16.06	−26.62	−36.69
	South Korea	−2.86	−7.28	−14.44	−18.32

MET, meteorological; EMIS, emission; unit (%).

from OBS–MET show that the various emission reduction policies implemented in both countries have worked well to improve air quality (Zhang et al., 2022a; Wang et al., 2019). This implies that the actual abatement policy effectiveness has been achieved beyond what is indicated by observations, and the meteorological effects should be added and corrected for.

3.5. Impact of emission reduction policies

To understand the impact of emission reduction policies in the past 5 years on air quality in China and South Korea, two types of emission data described in Section 2.2 were used to simulate the CMAQ model. The impact of emission reductions for 2017–2020 compared with that for 2016 were calculated through the model simulations (Table 2(b)).

In China, $PM_{2.5}$ concentrations have decreased annually due to emission control measures and changes have been recorded in the range of −3.49 % to −36.69 % from 2017 to 2020, compared with that in 2016. Similarly, South Korea showed the same trend as China, even though the rate of decrease (−2.86 % to −18.32 %; 2017–2020) was lower than that of China.

3.6. Impact of unexpected events on air quality in 2020

The coronavirus pandemic (COVID-19) began at the end of 2019 and has significantly impacted socio-economic activities worldwide (Chakraborty and Maity, 2020; Bashir et al., 2020). Previous studies (Cadotte, 2020; Chen et al., 2021; Krecl et al., 2020; Li et al., 2020; Manan et al., 2020) attempted to identify the consequential changes in the atmospheric environment due to several kinds of non-pharmaceutical interventions (i.e., lockdown, social distancing, etc.), which have been implemented by governmental authorities since early 2020 to prevent wide-spread of the virus. Moreover, in South Korea, special winter season countermeasures against high PM concentrations were introduced in early 2020 and implemented with significant governmental authority (Son et al., 2020), while a similar policy for emission reduction during the winter season has already been implemented in China since 2017.

In this study, we separated the impact of these unique but unexpected events (i. e., COVID-19 and the newly introduced reduction measures in 2020) on $PM_{2.5}$ concentrations, especially during winter 2020, from several other factors including meteorological changes and long-term planned emission reductions. The model simulation was conducted with the year-to-year adjusted emission during winter 2016–2019 to obtain the tendency of changing $PM_{2.5}$ concentrations, thereby reflecting the rate of change before the pandemic. By assuming that the existing long-term efforts for emission reduction would continue, and extending the slope onto 2019, the extrapolated concentrations during winter 2020 can be obtained. This would be the concentrations without considering any other unexpected reduction efforts or events, including COVID-19 measures. The difference between the extrapolated concentrations and the actual observed concentrations during winter 2020 will then likely be the result of unexpected events in 2020 (COVID-19 in China; COVID-19 and additionally implemented 2020 winter special measures in South Korea).

In China, the extrapolated concentration (51.24 μg/m^3) during winter 2020 was calculated to be approximately 3.13 μg/m^3 higher than the observed extrapolated concentration (48.11 μg/m^3), indicating that the $PM_{2.5}$ concentrations might have decreased by 3.13 μg/m^3 due to the contribution of pandemic measures to emission reduction. It is expected that the difference (5.73 μg/m^3) between the extrapolated (29.45 μg/m^3) and observed (23.72 μg/m^3) concentrations in Korea would be much larger than that in China, because the difference between the extrapolated and observed concentrations in South Korea are supposed to not only depend on COVID-19 measures, but also on newly introduced special measures during winter 2020. Unfortunately, it is beyond the scope of this study to separate the effect of COVID-19 measures from the combined contribution, which showed an approximate 20 % decrease in $PM_{2.5}$ concentrations in South Korea.

3.7. Estimated contribution of each impact factor to the total changes in air quality

We investigated the factors that have affected the air quality of China and South Korea over the past 5 years and explained how much they have contributed to the significantly decreased $PM_{2.5}$ concentrations during winter 2020 compared to that in 2016. The possible factors that contributed to the change in $PM_{2.5}$ concentration were divided into different year-to-year meteorological conditions, emission reduction effects, and impact of unexpected events, such as the COVID-19 pandemic and the newly implemented seasonal countermeasures during winter 2020. How the contributions were calculated is schematically illustrated in Fig. 5, and the quantitative contribution of each impact factor is shown in Fig. 6, which also includes numerical values for each contribution.

The observed $PM_{2.5}$ concentration of China was 62.80 μg/m^3 and 48.11 μg/m^3 during winter 2016 and 2020, respectively, and it has decreased by approximately −23.4 % over the past 5 years. As mentioned in Section 3.4, the contribution of the meteorological impacts can be driven by calculating the difference between MOD_{E2016}^{M2016} and MOD_{E2016}^{Myyyy} by fixing emission to 2016 and changing only the year-to-year meteorology. If the calculated rate of change ($[slope]_M$; the annual tendency depending on Table 2a) is applied into the observed $PM_{2.5}$ concentration of China during winter 2016, the expected $PM_{2.5}$ concentration ($[PM_{2.5}]_M$) of China during winter 2020 would be 67.56 μg/m^3 due to only meteorological impacts, which increased by +7.6 % (+4.77 μg/m^3) compared to the concentration of $PM_{2.5}$ during winter 2016. The results of the simulation with the adjusted emission amounts showed that the extrapolated concentration ($[PM_{2.5}]_E$) of China during winter 2020 was 51.24 μg/m^3, which has decreased by −26 % (−16.32 μg/m^3; $[PM_{2.5}]_E - [PM_{2.5}]_M$) during winter 2020, as compared with that in 2016. Thus, the long-term planned emission reduction policy would have contributed to the decreased concentrations, and any other unexpected reduction efforts or events including COVID-19 measures were not included, as mentioned in the previous section. Additionally, in China, there was a difference between the observed $PM_{2.5}$ concentrations ($[PM_{2.5}]_O$; 48.11 μg/m^3) and the extrapolated concentrations ($[PM_{2.5}]_E$) during winter 2020, which can be assumed to be the impact of lockdown and social distancing during the COVID-19 pandemic. The difference between $[PM_{2.5}]_E$ and $[PM_{2.5}]_O$ was −3.13 μg/m^3, indicating that the concentration of $PM_{2.5}$ in 2020 decreased by approximately −5 % compared with that in 2016 due to the influence of COVID-19 countermeasures in China.

Similarly, the contribution of each factor that impact the air quality in South Korea was examined. The observed $PM_{2.5}$ concentration in South Korea was 29.45 μg/m^3 and 23.72 μg/m^3 in 2016 and 2020, respectively, which decreased by approximately −19.5 % in 2020 compared with that in 2016. The $[PM_{2.5}]_M$ concentration was estimated to be 32.32 μg/m^3; thus, the $PM_{2.5}$ concentration during winter 2020 increased by approximately +9.7 % (+2.87 μg/m^3) compared with that in 2016 due to the different meteorological conditions. The difference, $[PM_{2.5}]_E - [PM_{2.5}]_M$, can explain the contribution of the existing long-term planned emission reduction efforts of Korea. This difference showed a decrease of −9.1 % (−2.69 μg/m^3) during winter 2020, as compared with that in 2016. Two unexpected events in Korea during winter 2020, i.e., social distancing and the newly introduced special reduction policy, also made a significant contribution to the decreased PM concentrations during winter 2020. The difference between $[PM_{2.5}]_O$ and $[PM_{2.5}]_E$ can reflect the combined

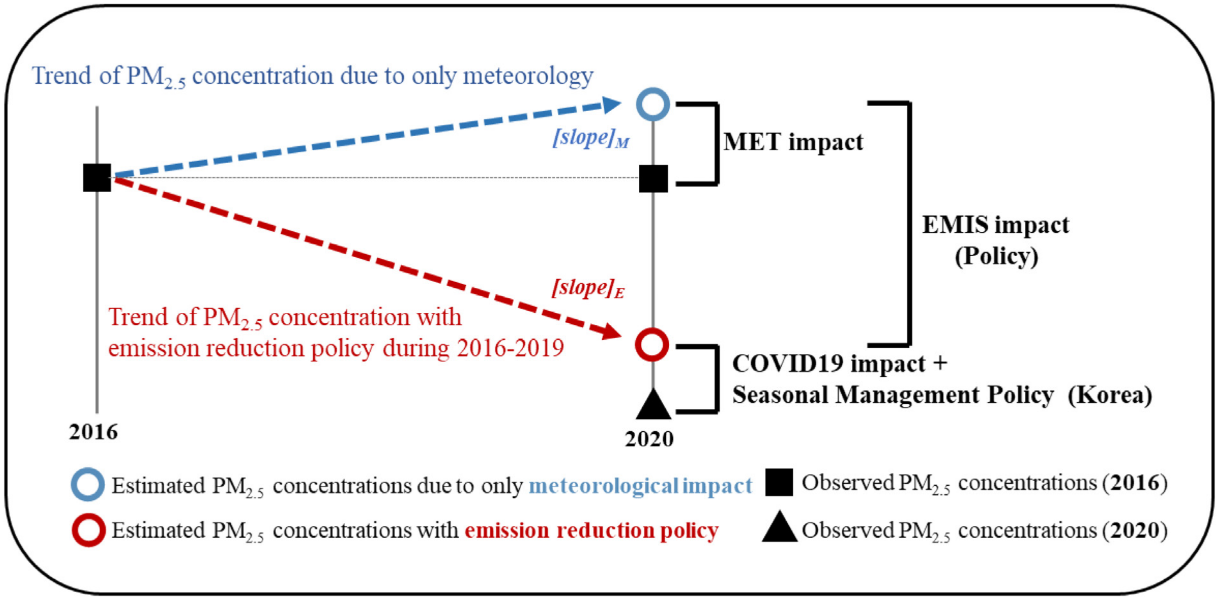

Fig. 5. Schematic diagram of the method used to analyze the contribution of factors affecting air quality.

contributions of these two unexpected events during winter 2020 in Korea, which was calculated to be −5.92 μg/m^3 (−19.5 % decrease compared with that in 2016).

Furthermore, attribution analysis was also conducted for metropolitan regions of both China and Korea using the same methodology, as it would allow us to identify differentiating features and special attentions in large cities (Liu et al., 2019; Wang et al., 2018; Chae, 2010). In Fig. 6, we showed the results of the attribution analysis for the BTH (Beijing, Hebei, Tianjin) region in China and the SMA (Seoul Metropolitan Area) in South Korea. The observed $PM_{2.5}$ concentrations of BTH region were 86.66 μg/m^3 and 63.17 μg/m^3 during winter 2016 and 2020, respectively, and it has decreased by approximately −27.1 % over the past 5 years. Applying the same methodology over BTH region, the results showed that the $PM_{2.5}$ concentration increases about +15.74 μg/m^3 (+22.2 %) due to meteorological effects was about three times higher than in whole of China (+7.6 %), indicating that the BTH region is more vulnerable to meteorological impacts than other Chinese regions. The decrease of $PM_{2.5}$ concentrations (−18.81 μg/m^3; −26.5 %) due to the long-term planned emission reduction policy was almost the same as in the nationwide (−26 %), and the decrease due to the outbreak of COVID-19 in 2020 was approximately −4.68 μg/m^3 (−6.6 %) which might be comparable to the decreasing amount per a year under the long-term policy (−4.7 μg/m^3), indicating a significant impact. For the SMA, the observed $PM_{2.5}$ concentrations was 32.25 μg/m^3 in 2016 and 26.18 μg/m^3 in 2020, indicating a reduction of approximately −18.8 % between 2016 and 2020. The meteorological impact was 2.91 μg/m^3 (+9 %) almost as the same as that over the entire country (+9.7 %), and the decrease in $PM_{2.5}$ concentrations due to long-term planned emission reduction efforts was 3.99 μg/m^3 (−13.6 %), which was greater than the nationwide (−9.1 %). This is likely to reflect the effect of prioritizing air pollution measures in the special management region, such as the Seoul Metropolitan Area. SMA region, where automobiles are the main source of emissions, shows a lower decreasing rate (−7.1 %) due to COVID-19 and seasonal special management policy in 2020 rather than the nationwide, where point sources such as industrial facilities are mainly located in. Nevertheless, the impact of unexpected events in 2020 over the SMA (−2.1 μg/m^3) can still be seen as significant in that it is twice as large as the decreasing (−1 μg/m^3 per a year) driven by long-term reduction policies.

We can also simply convert the accumulated contribution during 2016–2020 into the change per year (Fig. 6). The result shows that the $PM_{2.5}$ concentrations in China increased by +1.19 μg/m^3 annually (2016–2020) due to meteorological impacts, decreased by −4.08 μg/m^3 annually due to the existing long term planned emission control policy, and decreased by −3.13 μg/m^3 during winter 2020 due to the impact of COVID-19 countermeasures. In the case of BTH, the impacts of meteorological conditions, long-term planed emission control policies, and COVID-19 has been represented annually as +3.49 μg/m^3, −4.70 μg/m^3, −4.68 μg/m^3, respectively. In particular, the decrease in $PM_{2.5}$ concentrations due to the impact of COVID-19 has been found to be almost equivalent to the policy-driven reduction annually in $PM_{2.5}$ concentrations, which can be considered a significant impact.

Considering South Korea, the contribution of the meteorological impacts appears to have resulted in a + 0.72 μg/m^3 increase annually. The existing long term planed emission control policy may have contributed to a − 0.67 μg/m^3 decrease annually, and the two unexpected events (social distancing and the newly introduced special reduction policy) contributed to a − 5.92 μg/m^3 decrease during winter 2020. Annually, the impacts of meteorological conditions, long-term planed emission control policies, and unexpected events in 2020 have been indicated as +0.73 μg/m^3, −1.0 μg/m^3, and − 2.08 μg/m^3 respectively in SMA. The decreasing in $PM_{2.5}$ concentrations attributed to winter season special reduction policy and COVID-19 was found to be less significant in SMA compared to the nationwide over Korea. This may be explained with the difference of major emission sources between SMA and the nationwide; mainly mobile sources in SMA vs. point sources such as industrial facility in the rest region over Korea (Kim and Lee, 2018).

4. Discussion and conclusions

The factors that affected the air quality of China and South Korea during 2016–2020 were examined in this study. To assess the quantitative contribution of each factor, CMAQ model simulations and numerical experiments were conducted based on several scenarios. Based on the observation data, temporally and spatially adjusted emission data were provided as the input data of CMAQ model.

Surface observation data from China and South Korea were used to analyze the change in $PM_{2.5}$ concentrations during winter 2016–2020. The results showed that the concentration of $PM_{2.5}$ during winter was much higher than that of other seasons, and high concentration events occurred more frequently. The mean concentration of $PM_{2.5}$ during winter continued to decrease from 2016 to 2020 by −23.4 % and − 19.5 % in China and South Korea, respectively.

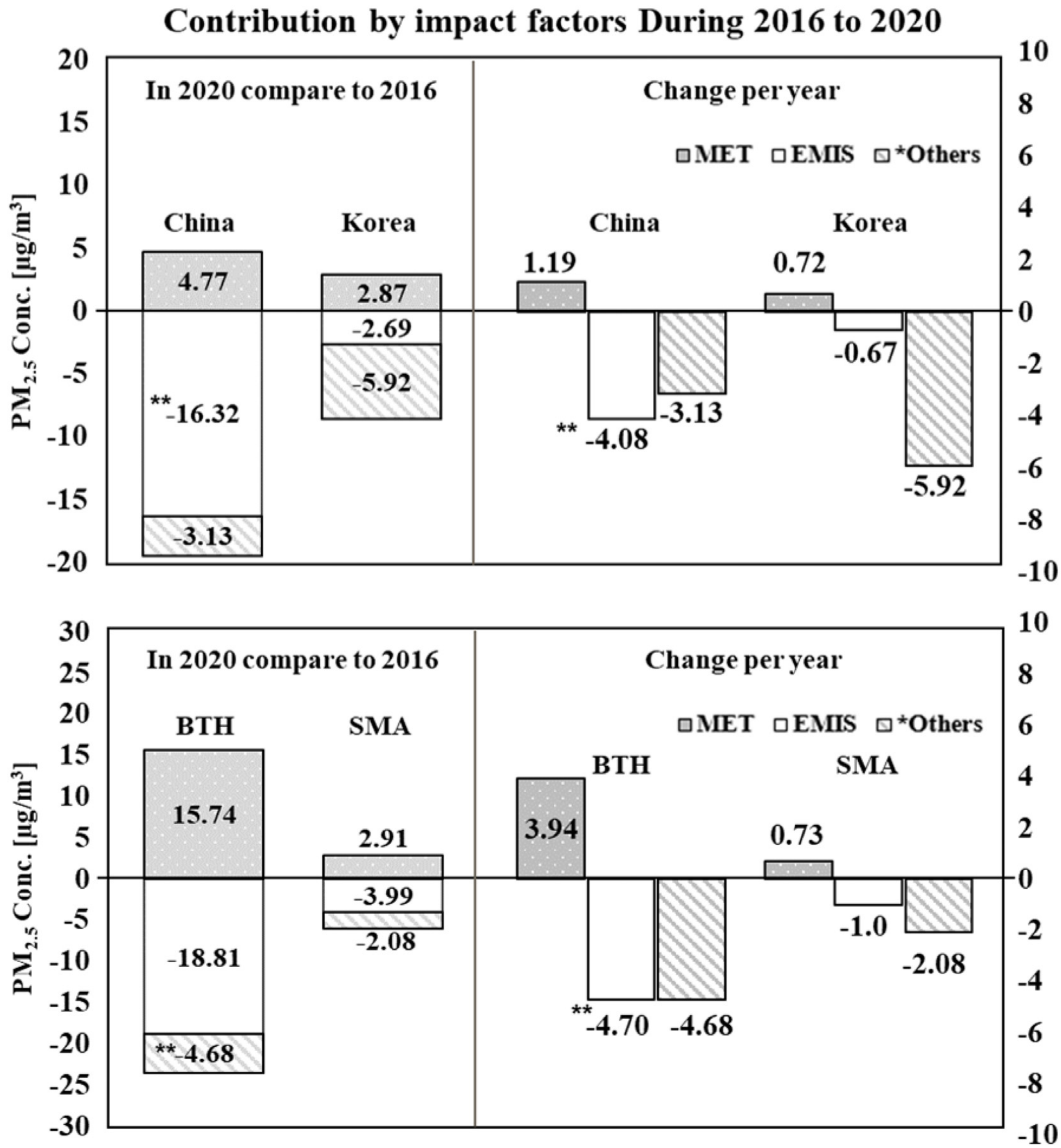

Fig. 6. Analysis of contribution by factors impacting air quality and the annual change in $PM_{2.5}$ concentrations.
*Others: Including the impact of COVID-19 (China, Korea, BTH, SMA) and the winter season special reduction policy newly implemented in 2020 (Korea, SMA).
** In China and BTH, Emission Reduction Policy include both Long Term and Seasonal Management Policy.

From the results of the CMAQ model simulations that applied two types of emission (KORUSv5, Adjusted KORUSv5), it was established that the concentration of $PM_{2.5}$ in China during winter 2020 increased by +7.6 % (+4.77 μg/m^3) compared with that in 2016 due to different meteorological conditions, decreased by −26 % (−16.32 μg/m^3) due to the existing long-term planned emission reduction measures, and further decreased by −5 % (−3.13 μg/m^3) due to the impact of the unexpected events during winter 2020, which included countermeasures against the COVID-19 pandemic. By converting total contribution during 2016–2020 into the annual change, the decreased annual concentration by −4.08 μg/m^3 due to the long-term emission control policies was found to be quite comparable to the −3.13 μg/m^3 decrease in only winter 2020 due to the impact of COVID-19 countermeasures. It is evident that South Korea has also experienced a similar change in the $PM_{2.5}$ concentration during winter 2016–2020. The meteorological impact appears to result in a + 0.72 μg/m^3 increase annually, while the existing long term planned emission control policy resulted in a − 0.67 μg/m^3 decrease. These kinds of significant features were also obviously but differently shown in the major cities of China and South Korea, such as BTH and SMA, respectively. The impact of meteorological changes was not negligible, and the policymaker should consider these additional contributions due to climatological changes to establish a long-term target for improved air quality. In South Korea, there were two different unexpected events that affected the change in concentration during winter 2020; i.e., social distancing and the newly introduced special reduction policy. The combined contribution of these two events resulted in a − 5.92 μg/m^3 decrease during winter 2020.

In this study, we were able to make a comprehensive assessment to quantify the contributions of possible factors to the recent change in air quality in both China and South Korea from 2016 to 2020. It is expected that the findings of this study may help the policymakers get the scientific basis to provide the relevant measures to improve the air quality in East Asia region.

CRediT authorship contribution statement

Yesol Cha: Result interpretation and writing – original draft preparation.
Chang-Keun Song: Supervision and leadership, and writing – review & editing.
Kwon-ho Jeon: Resources and investigation.
Seung-Muk Yi: Methodology, result interpretation and data analysis.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.163524.

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