



## Influence of Different Foreign Emissions Inventories on Simulated, Ground-Level Ozone in the Seoul Metropolitan Area during May 2014

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### ABSTRACT

This study examines the effects of different foreign anthropogenic emissions inventories on predicted ozone concentrations in the Seoul Metropolitan Area (SMA), South Korea, and estimates changes in ozone due to emissions reductions. We ran the Community Multi-Scale Air Quality (CMAQ) model using the High-Order Decoupled Direct Method with three inventories of foreign anthropogenic emissions: (1) the Intercontinental Chemical Transport Experiment, Phase B (INTEX-B) 2006; (2) the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) 2010; and (3) the Model Inter-Comparison Study (MICS)-Asia 2010. All three inventories have different spatial distributions of emissions, yielding different modeled ozone concentrations. However, the ozone concentrations modeled for the SMA differ less than those modeled for large, foreign cities in the modeling domain. The simulations using INTEX-B 2006 and CREATE 2010 suggested greater reduction in ozone with NO<sub>x</sub> control than with VOCs control. All simulations show that (1) simultaneous reduction in NO<sub>x</sub> and VOCs leads to less ozone reduction than the sum of ozone reductions for individual NO<sub>x</sub> and VOCs controls and (2) ozone reductions are stronger for high ozone days than for low ozone days. Comparing the modeled reductions in the relative sense yields smaller differences between high and low ozone days than comparing the modeled reductions in the absolute sense. With a 20% reduction in only NO<sub>x</sub> emissions, the differences in MDA1O3 among the three inventories were between 0.3 and 0.7 ppb. Because air-quality planning often leads to defined tonnage reductions, we examined the model's response to such a defined emissions reduction. Using the NO<sub>x</sub> reduction in China estimated by Zhao *et al.* (2013), we estimated that the differences in MDA1O3 among the three inventories were between 1.50 and 1.78 ppb. Based on these results, we recommend using different foreign anthropogenic emissions inventories to test future scenarios for air-quality control.

**Keywords:** Anthropogenic emissions; Ozone; Sensitivity; Seoul; Foreign inventory; CMAQ.

### INTRODUCTION

Ground-level ozone is a persistent air-quality problem in many areas around the world (World Health Organization, 2008; Royal Society, 2010; Wang *et al.*, 2012; U.S. EPA, 2013; Santurtún *et al.*, 2015). To be effective, local ozone abatement requires comprehensive planning due to the non-linear and complex characteristics of atmospheric ozone formation. Problems with ozone can be attributed both to local precursor emissions and to photochemical reactions

producing ozone (Seinfeld and Pandis, 2016), as well as to the regional transport of ozone formed in upwind areas (West *et al.*, 2009; Stock *et al.*, 2013). Precursors can also form ozone locally after being transported into an area. From a scientific point of view, controlling ozone is chemically challenging because the chemistry of ozone formation has non-linear characteristics; for instance, unlike concentrations of primary air pollutants, such as black carbon, ozone concentrations do not change linearly in proportion to changes in precursor emissions (Cohan *et al.*, 2005). The nonlinearity of ozone chemistry has been well-observed both nearby and far away from intensive precursor sources, such as urban areas with heavy traffic and power plants (Sillman, 2000; Ryerson *et al.*, 2001). From the point of view of the development of air-quality policy, controlling ozone is also

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challenging because upwind sources may lie outside the political or administrative boundaries of an area experiencing ozone air-quality issues, therefore falling outside the jurisdiction of that area's environmental authority (Farrell and Keating, 2002). Thus, air-quality improvement requires effective, reliable control strategies that consider not only the contributions of local and remote emissions sources but also non-linear chemistry and jurisdictional issues. In addition, it is quite challenging to develop scientifically defensible air-quality plans, since such plans rely on inputs from tools with a certain degree of uncertainty (Russell and Dennis, 2000). Among all inputs to air-quality models, emission inventories have often been noted as the major source of uncertainty (Fine *et al.*, 2003; Digar *et al.*, 2011).

In South Korea, for domestic anthropogenic emissions, most modeling studies use the Clean Air Policy Support System (CAPSS) developed and maintained by the National Institute of Environmental Research (National Institute of Environmental Research, 2015). For domestic and foreign biogenic emissions, the Model of Emissions of Gases and Aerosols from Nature (MEGAN) is most frequently used in South Korea (Guenther *et al.*, 2006). For foreign anthropogenic emissions in East Asia, however, three emissions inventories are most commonly used: (1) the Intercontinental Chemical Transport Experiment, Phase B (INTEX-B) 2006 (Zhang *et al.*, 2009); (2) the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) 2010 (Woo *et al.*, 2014); and (3) the Model Inter-Comparison Study (MICS)-Asia 2010 (Li *et al.*, 2017).

Prior work has studied the influence of emissions of nitrogen oxide ( $\text{NO}_x$ ) and volatile organic compounds (VOCs) from foreign countries on surface ozone concentrations in South Korea (Itahashi *et al.*, 2013; Bae *et al.*, 2014; Choi *et al.*, 2014), finding that ozone in Northeast Asia and South Korea has multiple aspects. The Northeast Asia region is more sensitive to  $\text{NO}_x$  than to VOCs, while springtime Chinese ozone can make significant contributions to ozone in neighboring countries. Increased Chinese  $\text{NO}_x$  emissions may lead to elevated ozone concentrations in the SMA, and mid-latitude regions of China may make large contributions to ozone in South Korea. However, these prior works examining the impact of foreign emissions on South Korean ozone are limited in terms of the number of foreign emissions inventories selected for direct comparison (often only one) or focus narrowly on the contribution assessment without exploring sensitivity of ozone to precursors. Assuming there is no clear answer about which foreign anthropogenic emissions inventory in East Asia is the most accurate, South Korean air-quality planners have difficulty in developing domestic air-quality improvement plans because the estimated effectiveness of control strategies may depend on choices about foreign emissions inventories. At a minimum, understanding the ozone-response characteristics of the results modeled using various foreign emissions inventories should help South Korean air-quality planners assess the effectiveness of control strategies by allowing planners to include such characteristics as part of their "weight of evidence" (U.S. EPA, 2007). Therefore, we attempted to

compare simulated local ozone concentrations and characterize ozone responses to changes in foreign emissions using multiple inventories of foreign anthropogenic emissions: INTEX-B 2006, CREATE 2010, and MICS-Asia 2010.

In detail, our approach involved the following. First, we simulated ozone with the three foreign emissions inventories, assessing model performance by comparing simulated and observed ozone concentrations in the Seoul Metropolitan Area (SMA), South Korea. Second, we used the sensitivity coefficients obtained with the High-Order Decoupled Direct Method (HDDM; Hakami *et al.*, 2003) to calculate the changes in ground ozone concentration in the SMA due to changes in foreign emissions, testing all three foreign emissions inventories. Implementing emissions controls, especially for large sources, such as the installation and operation of  $\text{NO}_x$ -control devices, often leads to defined reductions in the tonnage of emissions rather than percentage reductions relative to total emissions. Thus, rather than performing a typical sensitivity analysis using the ratio of emissions changes to total emissions, we examined the impact of planned, defined reductions in the tonnage of foreign anthropogenic emissions on ozone concentrations in the SMA, South Korea.

## MODEL SETUP

For this study, we used the Integrated Multidimensional Air-Quality System for Korea (IMAQS/K; Kim *et al.*, 2015), which comprises multiple versions of the Weather Research and Forecasting (WRF) meteorological model, the Sparse Matrix Operator Kernel Emissions (SMOKE) processing system, and the Community Multi-Scale Air Quality (CMAQ) photochemical grid model (Kim, 2011). IMAQS/K also runs various sensitivity and diagnostic tools, including the HDDM. For this work, we used an ensemble member of the IMAQS/K system with CMAQ-HDDM. In later sections, we introduce the HDDM and explain how we used it.

Modeling system inputs and configuration used for this study are as follows. Details of the WRF and CMAQ configurations are listed in Tables 1 and 2, respectively, adopted based on previous work regarding daily operational forecast modeling (Kim, 2011). For meteorological inputs to CMAQ, we used outputs of an IMAQS-K's WRF member using initial conditions and boundary conditions generated from the Global Forecasting System results (<https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forecast-system-gfs>) with no nudging. Fig. 1 shows the 27-km CMAQ modeling domain and the HDDM region in which modeled foreign anthropogenic emissions are reduced for this study. Choice of horizontal grid may significantly impact study results. Previous studies of regional transport in Northeast Asia have used 80-km (Itahashi *et al.*, 2013) and 60-km (Choi *et al.*, 2014) resolution to assess the impact of remote sources on local ozone air quality. Past studies in other regions have indicated that a 27-km resolution may suffice for a transport study focusing on upwind impacts (Cohan *et al.*, 2006; Wild and Prather, 2006; Ito *et al.*, 2009;

**Table 1.** WRF configuration.

| WRF Configuration               | Option                         |
|---------------------------------|--------------------------------|
| Version                         | 3.5.1                          |
| Cloud Microphysics              | WRF Single-Moment, Six-Class   |
| Shortwave Radiation             | Dudhia                         |
| Sub-grid Cumulus Scheme         | Kain-Fritsch                   |
| Planetary Boundary Layer Scheme | Yonsei University              |
| Longwave Radiation              | Rapid Radiation Transfer Model |
| Land Surface Model              | Noah                           |

**Table 2.** CMAQ configuration used in this study.

|                               |                          |
|-------------------------------|--------------------------|
| CMAQ Version                  | 4.7.1                    |
| Chemical mechanism            | SAPRC99                  |
| Aerosol                       | AERO5                    |
| Horizontal/Vertical Advection | YAMO                     |
| Horizontal Diffusion          | EDDY                     |
| Gas-phase Chemical Solver     | Euler Backward Iterative |

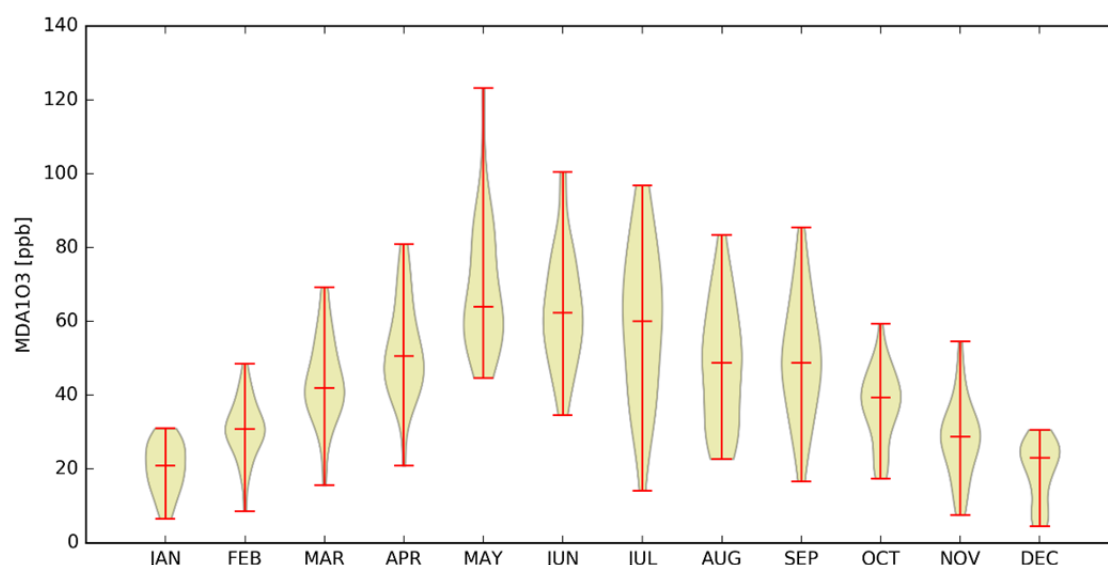


**Fig. 1.** 27-km CMAQ modeling domain and the SMA analysis area (inset). The shaded area represents the HDDDM source region, where foreign anthropogenic emissions are manipulated. The dots in the inset map show the location of ambient ozone monitors.

Schaap *et al.*, 2015). In Fig. 1, the dots in the right-hand panel represent the locations of the ambient ozone monitors, while the four-square boxes in the inset map depict the four, 27-km grid cells representing the SMA during analysis of the

impacts of changes in foreign anthropogenic emissions.

Fig. 2 presents a “violin plot” (Hintze and Nelson, 1998) of spatially averaged, maximum daily one-hour ozone concentrations (MDA1O3) measured at 81 monitors in the



**Fig. 2.** Spatially averaged MDA1O3 measured at 81 monitors in the SMA from January 1 to December 31, 2014. The width of each monthly plot represents the distributions of ozone concentration.

SMA from January 1 to December 31, 2014. The width of each monthly plot represents the frequency distributions of the measured ozone concentrations. In May 2014, the SMA experienced many high ozone days; given the significance of high ozone days for policymaking, we chose to model the period from May 1 to May 31, 2014 with a 10-day ramp-up beginning from April 21.

We used CAPSS 2011 for domestic anthropogenic emissions and MEGAN to estimate biogenic emissions. For foreign anthropogenic emissions, we used three emissions inventories: (1) INTEX-B 2006, (2) CREATE 2010, and (3) MICS-Asia 2010. Because their respective references offer detailed information about each foreign anthropogenic inventory, we summarize these inventories only briefly here. The INTEX-B 2006 emissions inventory was developed primarily to focus on anthropogenic emissions related to 22 countries in Northeast Asia, including China. INTEX-B 2006 is a gridded emissions inventory whose original spatial resolution is 0.5 degrees by 0.5 degrees. CREATE 2010 was developed by Woo *et al.* (2014) in South Korea for the Northeast Asia region based on Greenhouse Gas and Air Pollution Institute for Applied Systems Analysis (GAINS; IIASA, 2008; Klimont *et al.*, 2009). CREATE 2010 integrates existing emissions inventories for South Korea, North Korea, China, and Japan with emissions data for other Asian countries based on emissions comparisons by sector and activity (National Institute of Environmental Research, 2013). MICS-Asia 2010 resulted from recent collaborations between researchers from South Korea, China, and Japan (Li *et al.*, 2017). MICS-Asia 2010 is a gridded emissions inventory and it has spatial resolution of 0.25 degrees by 0.25 degrees. Table 3 summarizes the properties of the three foreign emissions inventories used in this study.

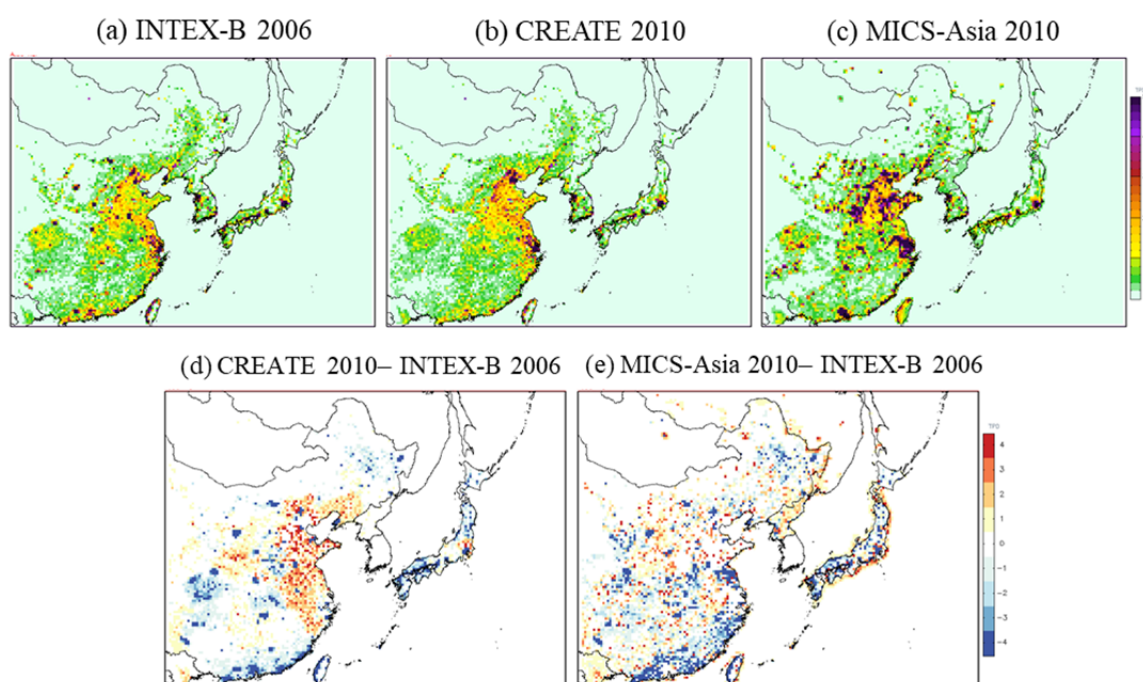
After preparing model-ready emissions input files, we examined these inputs' spatial characteristics. For  $\text{NO}_x$  emissions, Fig. 3 shows the spatial distribution and total daily average from INTEX-B 2006, CREATE 2010, and

MICS-Asia 2010 during the modeled period. We calculated spatial difference plots by subtracting the INTEX-B 2006 emission values from those of CREATE 2010 and MICS-Asia 2010, noting that the spatial differences in  $\text{NO}_x$  emissions between CREATE 2010 and INTEX-B 2006 are similar to those between MICS-Asia 2010 and INTEX-B 2006 for large cities in China. In many large Chinese cities, INTEX-B 2006 estimates much higher  $\text{NO}_x$  emissions than do the other two inventories. However, near Beijing, CREATE 2010 shows marginally higher  $\text{NO}_x$  emissions than INTEX-B 2006, while MICS-Asia 2010 has much larger  $\text{NO}_x$  emissions than does INTEX-B 2006 (and possibly CREATE 2010) in many other areas, likely those containing small or medium-sized cities and/or industrial complexes. Overall, the MICS-Asia 2010 inventory results in much larger total  $\text{NO}_x$  emissions than do the other two. For Chinese  $\text{NO}_x$ , INTEX-B 2006, CREATE 2010, and MICS-Asia 2010 estimate, respectively, 50,307 tons per day (TPD), 46,087 TPD, and 66,546 TPD, amounts that are 19 to 27 times South Korean  $\text{NO}_x$  emissions of 2,465 TPD. As shown in Table 3,  $\text{NO}_x$  emissions from the industrial sector in MICS-Asia 2010 is much larger than other two inventories while the transportation sector, including automobiles, shows large differences in  $\text{NO}_x$  between INTEX-B 2006 and other two inventories. For power plants, INTEX-B 2006 and MICS-Asia 2010 estimate comparable  $\text{NO}_x$  emissions. Therefore, we anticipated that the spatial distribution of the differences in modeled ozone concentration between MICS-Asia and the other two inventories would, at least, center on major industrial sources of  $\text{NO}_x$  emissions in China.

Fig. 4 shows the spatial distribution of VOCs emissions from INTEX-B 2006, CREATE 2010, and MICS-Asia 2010 over the modeled period. As with the  $\text{NO}_x$  emission comparison, INTEX-B 2006 served as the basis for comparison. The spatial differences in VOCs emissions between CREATE 2010 and INTEX-B 2006 are similar to those between MICS-Asia 2010 and INTEX-B 2006 in most

**Table 3.** Summary of the foreign anthropogenic emissions inventories used in this study.

|                                         | INTEX-B 2006                             | CREATE 2010                                                                                                              | MICS-Asia 2010                                                                       |
|-----------------------------------------|------------------------------------------|--------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|
| Type                                    | Gridded ( $0.5^\circ \times 0.5^\circ$ ) | Non-gridded                                                                                                              | Gridded ( $0.25^\circ \times 0.25^\circ$ )                                           |
| Temporal Scale                          | Annual                                   | Monthly                                                                                                                  | Monthly                                                                              |
| Inventory                               | Gaseous Species                          | CO <sub>2</sub> , CH <sub>4</sub> , N <sub>2</sub> O, CO, NO <sub>x</sub> ,<br>NMVOCs, NH <sub>3</sub> , SO <sub>2</sub> | CO <sub>2</sub> , CO, NH <sub>3</sub> ,<br>NMVOCs, NO <sub>x</sub> , SO <sub>2</sub> |
| Species                                 | Particulate Species                      | OC, BC, PM <sub>10</sub> , PM <sub>2.5</sub> , Hg                                                                        | EC, OC, PM <sub>2.5</sub> , PM <sub>10</sub>                                         |
| Chinese NO <sub>x</sub> Emissions (TPD) | 50,307                                   | 46,087                                                                                                                   | 66,546                                                                               |
| Industrial (TPD)                        | 13,053                                   | 16,168                                                                                                                   | 25,129                                                                               |
| Power plants (TPD)                      | 25,342                                   | 13,541                                                                                                                   | 22,919                                                                               |
| Transportation (TPD)                    | 11,912                                   | 16,231                                                                                                                   | 16,764                                                                               |
| Chinese VOC Emissions (TPD)             | 48,700                                   | 42,123                                                                                                                   | 48,712                                                                               |
| Industrial (TPD)                        | 28,681                                   | 33,720                                                                                                                   | 32,335                                                                               |
| Transportation (TPD)                    | 17,367                                   | 8,091                                                                                                                    | 8,015                                                                                |

**Fig. 3.** Daily average NO<sub>x</sub> emissions (TPD) in (a) INTEX-B 2006, (b) CREATE 2010, (c) MICS-Asia 2010, (d) CREATE 2010 minus INTEX-B 2006, and (e) MICS-Asia 2010 minus INTEX-B 2006 emissions inventories during May 2014.

of China, except for the northeast, near Manchuria. Overall, Chinese VOCs emissions in INTEX-B 2006 are much higher than those in CREATE 2010 and MICS-Asia 2010. However, near the border between China and Russia, as well as the border between China and Mongolia, Chinese VOCs emissions are smaller in INTEX-B 2006. Regarding Chinese VOCs in our study domain, INTEX-B 2006, CREATE 2010, and MICS-Asia 2010 estimated 48,700 TPD, 42,123 TPD, and 48,712 TPD, respectively, which are 15 to 17 times the estimated South Korean VOCs emissions of 2,865 TPD. As shown in Table 3, the transportation sector, including automobiles, shows large differences in VOCs between INTEX-B 2006 and other two inventories.

#### Sensitivity Modeling Approach

Sensitivity modeling is a technique for examining changes in output variables (e.g., O<sub>3</sub> concentrations),  $dO$ , due to

arithmetic changes in input variables (e.g., NO<sub>x</sub> emissions),  $d_I$ , as follows:

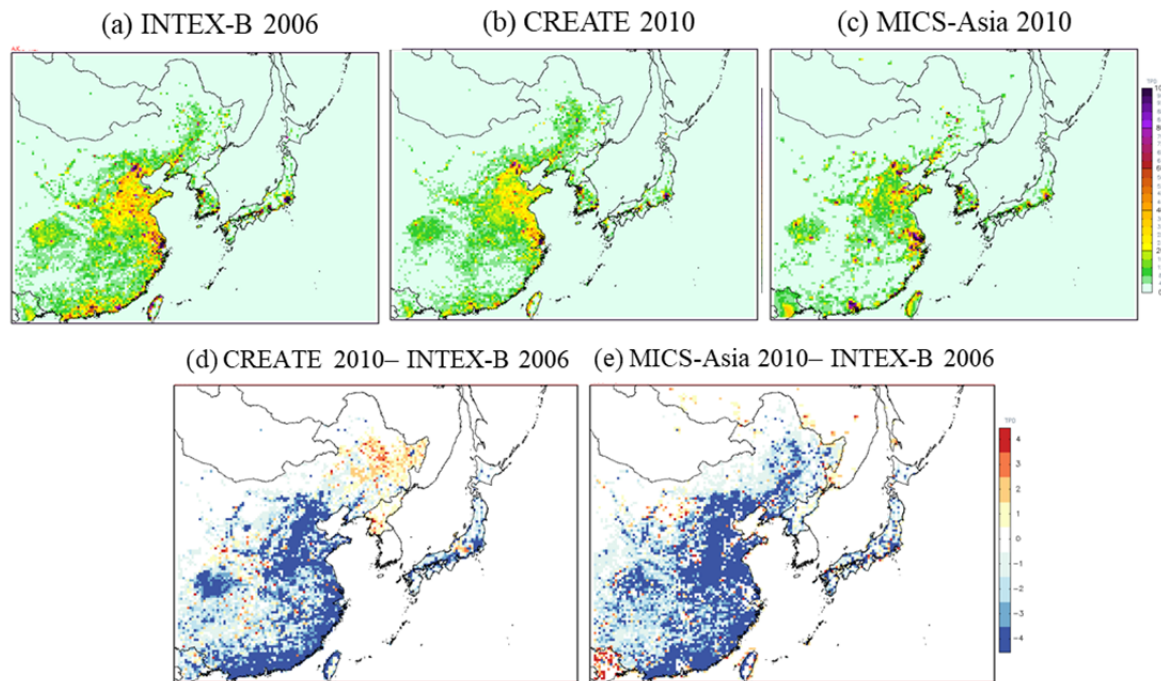
$$dO = O_{new} - O_{old} = f(I_{old} + d_I) - f(I_{old}) \quad (1)$$

where  $I_{old}$ ,  $O_{old}$ , and  $O_{new}$  represent, respectively, the input variable with no change and the output variables before and after applying changes to the input variable.  $f(x)$  is a function estimating air pollutant concentrations.

The Taylor series of  $O_{new}$  can be expressed as follows:

$$O_{new} = O_{old} + f'(O_{old})(d_I) + \frac{f''(O_{old})}{2}(d_I)^2 + Error \quad (2)$$

For modeling emissions sensitivity, the HDDM approximates changes in the modeled species concentrations



**Fig. 4.** Daily average VOCs emissions (TPD) in (a) INTEX-B 2006, (b) CREATE 2010, (c) MICS-Asia 2010, (d) CREATE 2010 minus INTEX-B 2006, and (e) MICS-Asia 2010 minus INTEX-B 2006 emissions inventories during May 2014.

with changes in emissions relative to the original emissions,  $r_I = (I_{new} - I_{old})/I_{old}$ , and estimates sensitivity coefficients up to the second derivative of Taylor series,  $S1$  and  $S2$  (Cohan et al., 2005). For modeling the sensitivity of an input variable, the equation for  $O_{new}$  above can be rewritten as follows:

$$O_{new} = O_{old} + S1(r_I) + \frac{S2}{2}(r_I)^2 + Error \quad (3)$$

where  $S1$  and  $S2$  are defined as  $\partial O / (\partial(I_{new}/I_{old}))$  and  $(\partial^2 O) / (\partial(I_{new}/I_{old})^2)$ , respectively.

For modeling ozone sensitivity to  $NO_x$  and VOCs, the equation can be expanded as follows:

$$O_{new} = O_{old} + S1_{NO_x}(r_{NO_x}) + \frac{S2_{NO_x}}{2}(r_{NO_x})^2 + S1_{VOCs}(r_{VOCs}) + \frac{S2_{VOCs}}{2}(r_{VOCs})^2 + S2_{NO_x \& VOCs}(r_{NO_x})(r_{VOCs}) + Error \quad (4)$$

where  $S2_{NO_x \& VOCs}$  is defined as  $\partial^2 O / (\partial(ENO_{x_{new}}/ENO_{x_{old}})\partial(EVOC_{new}/EVOC_{old}))$ .  $ENO_x$  and  $EVOC$  are  $NO_x$  and VOCs emissions, respectively.

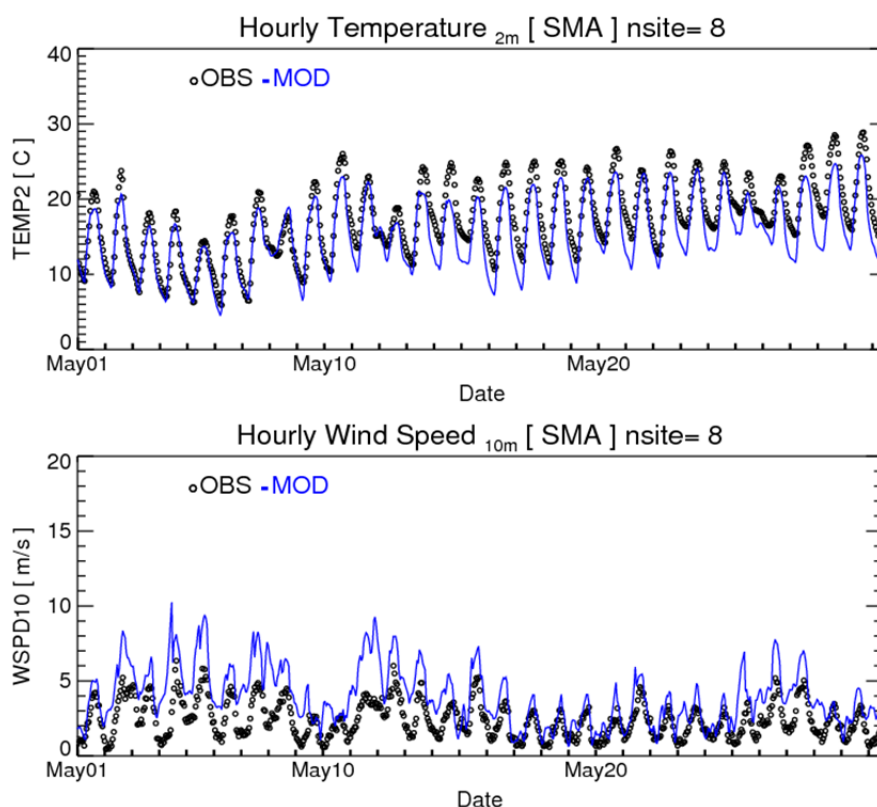
Here, the first-order sensitivity coefficient is the rate of changes in ozone concentration due to changes in emissions compared to total emissions in the base case. In other words, calculating actual changes in ozone using the sensitivity coefficient requires estimating changes in emissions normalized against the total emissions from the HDDM source region. The second-order sensitivity coefficient is

the rate of change of the first-order coefficient due to the ratio between emissions change and total emissions in the base case. Positive values of first-order sensitivity coefficients indicate that ozone in the area will increase with an emissions increase. Negative values of second-order sensitivity coefficients mean that the rates of change in ozone concentration due to emissions changes decrease, eventually becoming negative, if the decreasing tendency is maintained and the first-order sensitivity coefficient is initially positive. Previous studies have proven the validity of HDDM for a 20% reduction in emissions (Dunker et al., 2002; Yarwood et al., 2013). For  $NO_x$ , 20% reduced emissions based on the three foreign emissions inventories ranges from 9,217 to 13,309 TPD; for VOCs, 20% reduced emissions ranges from 8,425 to 9,742 TPD. Lastly, we multiplied the ratio of emissions change (e.g., 20%),  $r_I$ , by the sensitivity coefficients to estimate the impact of absolute changes in foreign anthropogenic emissions on ozone air quality in the SMA.

## RESULTS AND DISCUSSION

### Evaluation of Model Performance

First, we evaluated the performance of the WRF model during the modeled period. Fig. 5 compares spatially averaged 2-m temperatures and 10-m wind speeds at all Meteorological Assimilation Data Ingest System (MADIS) monitors in the 27-km domain during May. In general, the WRF model agrees well with observed 2-m temperatures, although small biases were observed during the evening hours on some days. For 10-m wind speeds, WRF also shows good agreement with observations on many days, especially near the end of May, although it has high biases during nighttime throughout the month. We also computed various meteorological model



**Fig. 5.** Time series of 2-m temperatures (top) and 10-m wind speeds (bottom) at all MADIS monitors in the 27-km domain during May 2014.

performance statistics for 2-m temperatures and 10-m wind speeds at the three Korea Meteorological Administration (KMA) sites in the SMA, as summarized in Tables 4 and 5. The meteorological station in Incheon is very close to shore (See Fig. S1), so the corresponding modeling grid cell has over 50% water fraction, which, we believe, heavily affects the estimation of meteorological variables at this cell. The modeled values may therefore be incomparable with observed values. Considering this, we conclude that the WRF model performs adequately for this study.

To examine transport patterns, we visualized surface- and 850 hPa-level winds and CO concentrations (in Figs. S2 and S3). We also conducted a series of HYSPLIT back trajectories, examining the series with hourly spatial distributions and vertical profiles of winds and CO concentrations (in Fig. S4). Overall, throughout May 2014, the prevalent surface-level winds were southwesterly and northwesterly in and around the SMA. We noticed frequent southerly winds over the Yellow Sea when the SMA was under stagnant conditions. CO was often transported from Shanghai to areas near the SMA during periods of high modeled ozone concentrations. At 850 hPa, winds also alternated southwesterly and northwesterly throughout the modeled period over the Yellow Sea and near the SMA. When relatively low wind speeds were modeled at the surface near the SMA, winds at 850 hPa were often westerly or stagnant. In terms of vertical transport heights, we noticed that most of the lower-level airmass in the SMA is likely affected by (or could be traced back to) East

Central or Southern China.

Fig. 6 compares the time series of observed and modeled MDA1O3 using the three foreign anthropogenic emissions inventories. Overall, the modeled ozone concentrations match the observed ozone concentrations well, with few differences among the modeled results resulting from all three emissions inventories, as scatter plots (Fig. 7) also confirm. The monthly average (minimum–maximum) values of MDA1O3 were 72 ppb (47–129 ppb), 74 ppb (46–133 ppb), and 73 ppb (48–138 ppb) from simulations using INTEX-B 2006, CREATE 2010, and MICS-Asia 2010, respectively, while the spatially averaged ozone concentration at the monitors was 69 ppb (45 ppb–123 ppb). Table 6 summarizes the performance statistics of all simulations for MDA1O3. Based on these visualizations and the performance statistics shown in Table 6 and Fig. 7, we concluded that the modeled results are reasonable for further analysis of sensitivities in the SMA to emissions reductions.

Figs. 8 and 9 show monthly mean, one-hour ozone concentrations and MDA1O3 for May 2014. Because of the large spatial variations in  $\text{NO}_x$  emissions, as shown in Fig. 3, we initially expected large spatial differences in modeled ozone concentrations. Overall, differences in the spatial distributions of ozone concentrations follow the spatial patterns of  $\text{NO}_x$  emissions. For example, in many large cities in China, the spatial pattern of ozone differences resembles that of  $\text{NO}_x$  in Fig. 3. Locations with larger  $\text{NO}_x$  emissions in the INTEX-B 2006 inventory tend to have lower ozone concentrations compared with those modeled

**Table 4.** Performance statistics for 2-m temperatures at the three KMA sites in the SMA during May 2014.

| Site Name          | Seoul | Incheon | Suwon |
|--------------------|-------|---------|-------|
| N                  | 744   | 744     | 744   |
| Mean MOD (degrees) | 15.8  | 13.1    | 16.0  |
| Mean OBS (degrees) | 18.9  | 16.8    | 18.2  |
| Bias (degrees)     | −3.1  | −3.7    | −2.2  |
| Error (degrees)    | 3.2   | 4.3     | 2.5   |
| RMSE (degrees)     | 3.8   | 5.2     | 3     |
| R <sup>2</sup>     | 0.8   | 0.3     | 0.9   |

where, N is the number of observation-model pairs.

$$\text{Mean Modeled (MOD)} = \frac{\sum P}{N},$$

$$\text{Mean Observed (OBS)} = \frac{\sum O}{N},$$

$$\text{Bias} = \frac{\sum (P - O)}{N}, \text{ Error} = \frac{\sum |P - O|}{N},$$

$$\text{Root Mean Square Error (RMSE)} = \sqrt{\frac{\sum (P - O)^2}{N}},$$

$$\text{Coefficient of Determination (R}^2\text{)} = 1 - \frac{\sum (O - P)^2}{\sum (O - \bar{O})^2},$$

P and O represent the modeled and observed values, respectively.

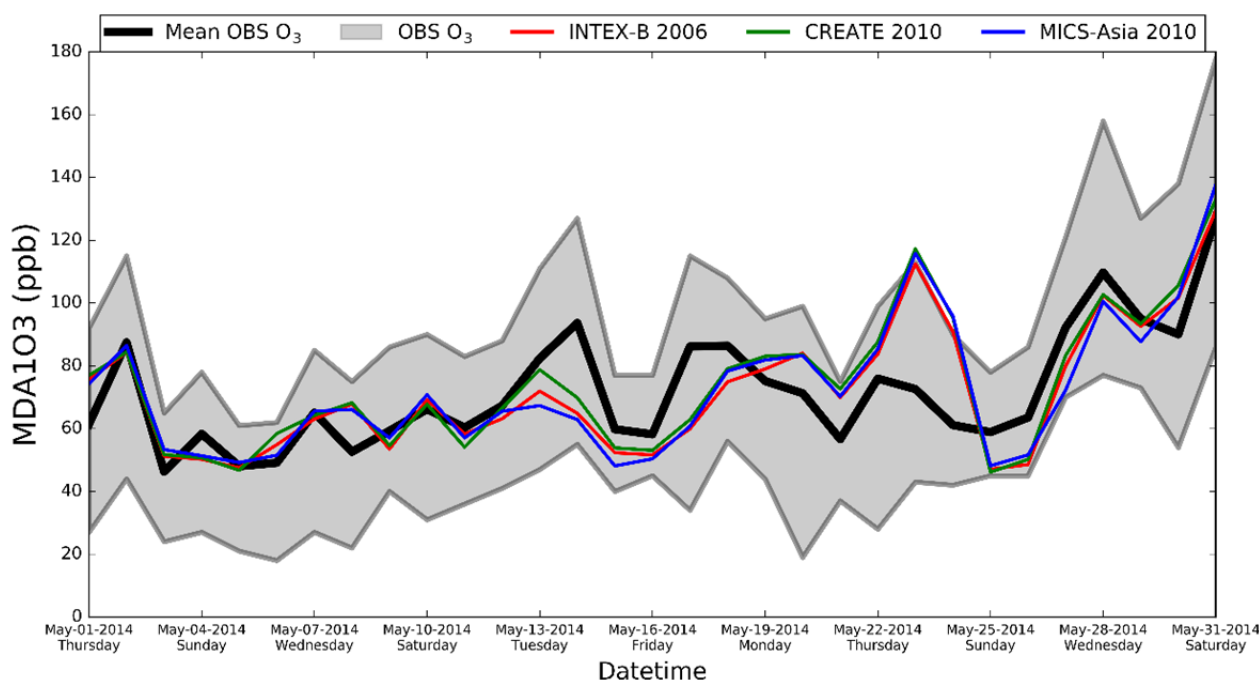
using CREATE 2010 and MICS-Asia 2010, which indicates significant NO<sub>x</sub> titrations due to excess NO<sub>x</sub>. Overall, MICS-Asia 2010 led to much more modeled ozone than did INTEX-B 2006 and CREATE 2010 in large cities. Over

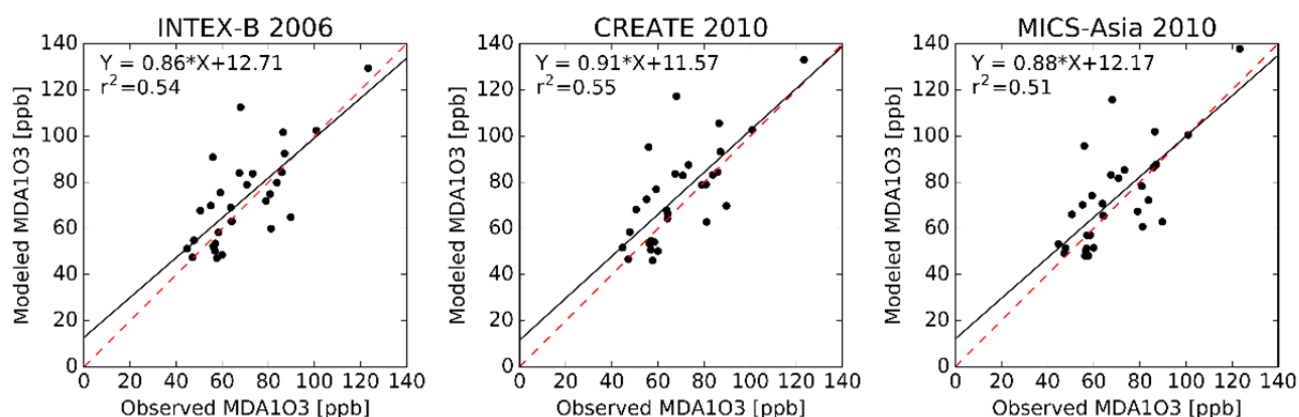
**Table 5.** Performance statistics for 10-m wind speeds at the three KMA sites in the SMA during May 2014.

| Statistics                    | Seoul | Incheon | Suwon |
|-------------------------------|-------|---------|-------|
| N                             | 744   | 744     | 744   |
| Mean MOD (m s <sup>−1</sup> ) | 3.2   | 5.3     | 3.3   |
| Mean OBS (m s <sup>−1</sup> ) | 3.2   | 3.2     | 2.1   |
| Bias (m s <sup>−1</sup> )     | 0.1   | 2.1     | 1.2   |
| Error (m s <sup>−1</sup> )    | 1.2   | 2.3     | 1.4   |
| RMSE                          | 1.5   | 2.9     | 1.8   |
| R <sup>2</sup>                | 0.4   | 0.4     | 0.5   |

the Yellow Sea, the differences between INTEX-B 2006 and CREATE 2010 for monthly average MDA1O3 were significantly larger than for monthly average, one-hour ozone concentrations. Such differences were not apparent between INTEX-B 2006 and MICS-Asia 2010. Meanwhile, the impact on the SMA was notably less pronounced than differences in local ozone concentrations in major areas of emission sources in China or over the Yellow Sea, which we inferred as evidence that apparent causal relationships between foreign emissions and surface ozone concentrations in the SMA were very weak during the modeled period.

However, interpreting this finding requires caution; the impact of emission differences from upwind sources may not be compared directly with the impact of domestic emissions. It is because dispersions including transport aloft and photochemical reactions also play important roles during airmass movement from upwind to downwind. For example, odd oxygen molecules that could contribute to ozone formation in upwind areas may be attached to nitrogen species (e.g., NO<sub>2</sub> or peroxyacetyl nitrate) depending on photochemical regimes in upwind areas and transported to

**Fig. 6.** Time series comparison of observed ozone and modeled MDA1O3 using the three foreign anthropogenic emissions inventories during the studied period.



**Fig. 7.** Spatial averages of observed ozone and modeled MDA1O3 using the three foreign anthropogenic emissions inventories during the studied period.

**Table 6.** MDA1O3 model-performance statistics of simulations using the three foreign anthropogenic emissions inventories.

|                | INTEX-B 2006 | CREATE 2010 | MICS-Asia 2010 |
|----------------|--------------|-------------|----------------|
| Mean OBS (ppb) | 68.8         |             |                |
| Mean MOD (ppb) | 72.1         | 74.1        | 72.5           |
| Bias (ppb)     | 3.4          | 5.3         | 3.8            |
| Error (ppb)    | 10.3         | 10.6        | 11.0           |
| RMSE (ppb)     | 14.3         | 15.3        | 15.4           |
| NMB (%)        | 4.9          | 7.7         | 5.5            |
| NME (%)        | 14.9         | 15.4        | 16.0           |
| FB (%)         | 3.9          | 6.3         | 4.3            |
| FE (%)         | 14.5         | 14.6        | 15.2           |
| R <sup>2</sup> | 0.54         | 0.55        | 0.51           |

$$\text{Normalized Mean Bias (NMB)} = 100\% \times \frac{\sum (P - O)}{\sum O}, \quad \text{Normalized Mean Error (NME)} = 100\% \times \frac{\sum |P - O|}{\sum O},$$

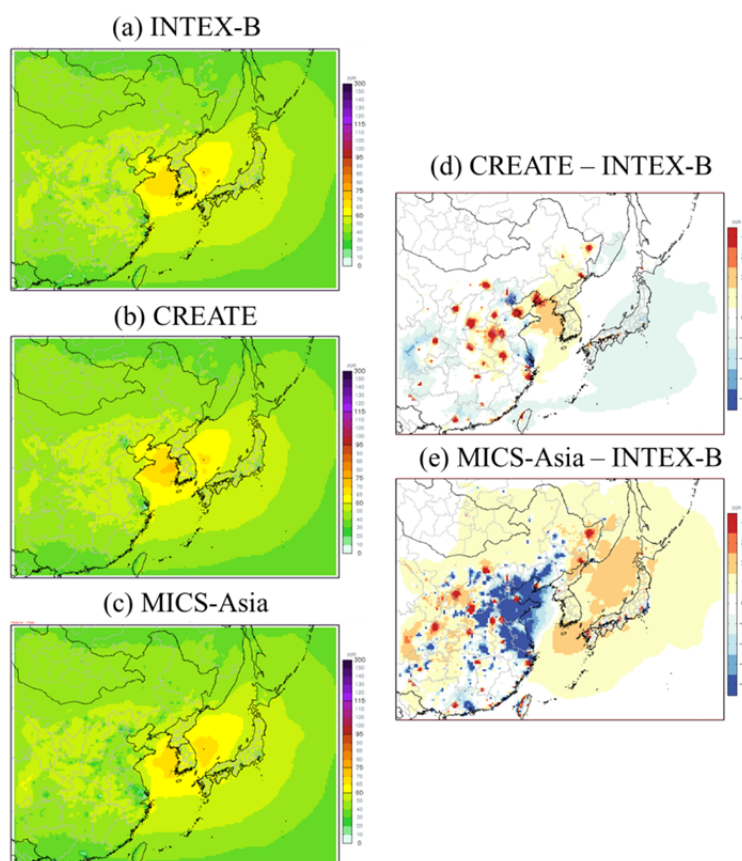
$$\text{Fractional Bias (FB)} = 100\% \times \frac{2}{N} \times \sum \frac{(P - O)}{(P + O)}, \quad \text{Fractional Error (FE)} = 100\% \times \frac{2}{N} \times \sum \frac{|P - O|}{(P + O)},$$

N, P, and O represent the number of observation-model pairs, predicted concentrations, and observed concentrations, respectively. For this table, N is 31 for all cases.

downwind areas. If downwind areas are under heavy NO<sub>x</sub> emission conditions, transported NO<sub>x</sub> may influence downwind ozone formation very little. In addition, the same amounts of different VOC species can produce different amounts of ozone, depending on photochemical characteristics. For this study, all inventories were processed using SAPRC99 for gas-phase chemistry and AERO5 for aerosol chemistry, as presented in Table 2. INTEX-B 2006 and CREATE 2010 have matched-source classification codes, while MICS-Asia 2010 does not. Therefore, we cross-referenced emissions sources between MICS-Asia 2010 and INTEX-B 2006. Considering domestic emissions and VOC speciation, further investigation will be needed to compare the relative impacts of foreign emissions.

Fig. 10 shows modeled, monthly average changes in ozone due to 20% reductions in NO<sub>x</sub> emissions only, VOCs emissions only, and both NO<sub>x</sub> and VOCs emissions for the three foreign emissions inventories. For reductions only in NO<sub>x</sub> emissions, INTEX-B 2006 and CREATE 2010 have similar spatial patterns. MICS-Asia 2010 led to much

larger positive increases in modeled ozone with 20% NO<sub>x</sub> reductions, especially near Beijing, Shanghai, and Hong Kong, which indicates that MICS-Asia 2010 tends to create a much more NO<sub>x</sub>-rich environment in the model compared with the other two inventories, eventually leading to less ozone reduction in the SMA. For reductions only in VOCs emissions, the three foreign emissions inventories produce similar spatial patterns, with the resulting ozone reduction in the SMA estimated around 1 ppb. When both NO<sub>x</sub> and VOCs emissions are reduced by 20%, INTEX-B 2006 and CREATE 2010 show similar spatial patterns, as in the NO<sub>x</sub>-only reduction case. Using MICS-Asia 2010, the model predicts about 1 ppb less ozone over South Korea. Interestingly, this inventory showed a clear transition between increasing and decreasing trends in ozone over the Yellow Sea. Because different synoptic weather patterns might bring that clear transition line nearer to or farther from the SMA, further study is needed under additional meteorological conditions. The implication is that the decrease in ozone concentration in the SMA estimated with



**Fig. 8.** Comparison of monthly mean, modeled one-hour ozone concentrations.

MICS-Asia 2010 may be much more susceptible to meteorological variability than that estimated with either of the other two foreign emissions inventories.

We calculated the variability of changes in modeled ozone concentration due to the choice of foreign emissions inventory. Ozone air-quality management often focuses on days over an administrative standard, which, in South Korea, is 0.1 ppm for one-hour ozone (<http://eng.me.go.kr/eng/web/index.do?menuId=253>). During May 2014, the simulations show, four days had one-hour ozone over 0.1 ppm. As Table 7 shows, all three cases with the different foreign anthropogenic emissions inventories modeled exceedances on the same four days.

For these four days, Fig. 11 shows the average modeled ozone changes due to 20% reductions in emissions: only  $\text{NO}_x$  reduction, only VOCs reduction, the sum of the only  $\text{NO}_x$  and only VOCs reductions, and both  $\text{NO}_x$  and VOCs reductions at once. The modeled results for all three foreign anthropogenic emissions inventories indicate that, when MDA1O3 is equal to or less than 100 ppb, the 20%  $\text{NO}_x$ -only control yields more ozone reductions than a 20% VOCs-only control, except for the model using the MICS-Asia 2010. For combined control of  $\text{NO}_x$  and VOCs, simultaneous controls notably yield less ozone reduction than the sum of the individual precursor controls, which implies that evaluating a control strategy must, in final implementation, account for ozone's sensitivity to simultaneous changes in its precursors, expressed as the cross-sensitivity term  $S2_{\text{NO}_x\&\text{VOCs}}$  in the

HDDM formula. Overall, high ozone days show stronger decreases in ozone concentration than do other days.

Because models are often used in a relative sense due to their uncertainties (U.S. EPA, 2007), we examined how models with different foreign anthropogenic emissions inventories show ozone responding to precursor emissions in a relative sense. Fig. 12 shows the models' relative responses to the precursor controls by status of air-quality standard exceedance. In a relative sense, the INTEX-B 2006 and CREATE 2010 inventories show larger responses to emission changes than MICS-Asia 2010 when MDA1O3 is lower than the air-quality standard. By contrast, MICS-Asia 2010 shows the opposite trend. All cases of high MDA1O3 show comparable responses to precursor control combinations. Descending order of the three emissions inventories by size of their relative modeled MDA1O3 response to precursor control combinations is (1) CREATE 2010, (2) INTEX-B 2006, and (3) MICS-Asia 2010, except for the 20% VOCs-only case, which shows similar responses for all three inventories. This finding suggests that choice of foreign emissions inventories does not create significant uncertainties when using modeled results primarily for high-ozone cases, especially in a relative sense. However, as discussed above, this may not hold true under different meteorological conditions, especially with MICS-Asia 2010.

Precursor controls are often defined by tonnage rather than as a reduction ratio, which could lead to different sizes or even trends of MDA1O3 changes compared with the

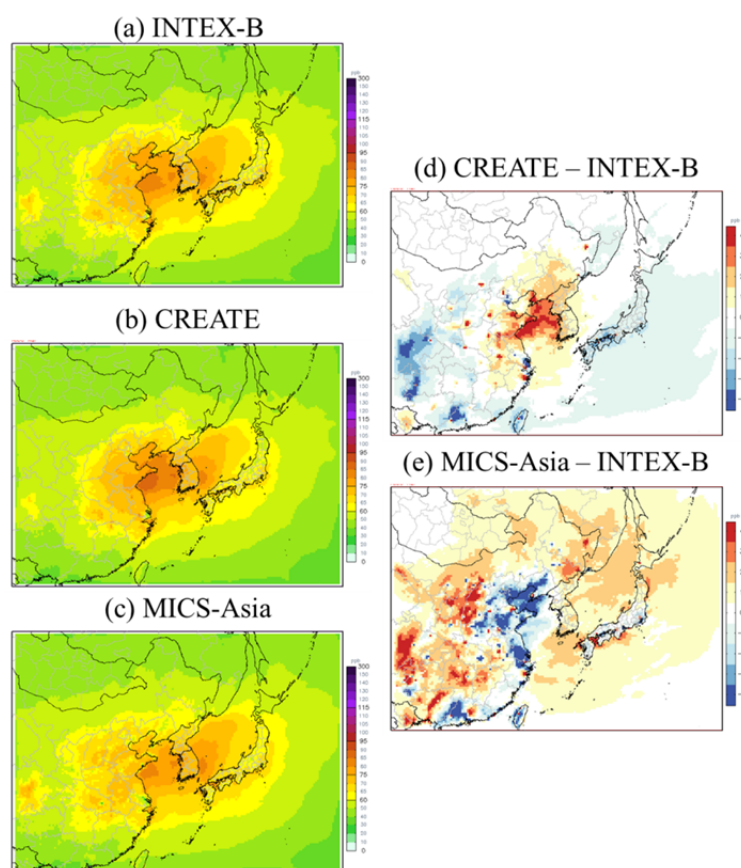


Fig. 9. Comparison of the monthly mean, modeled MDA103.

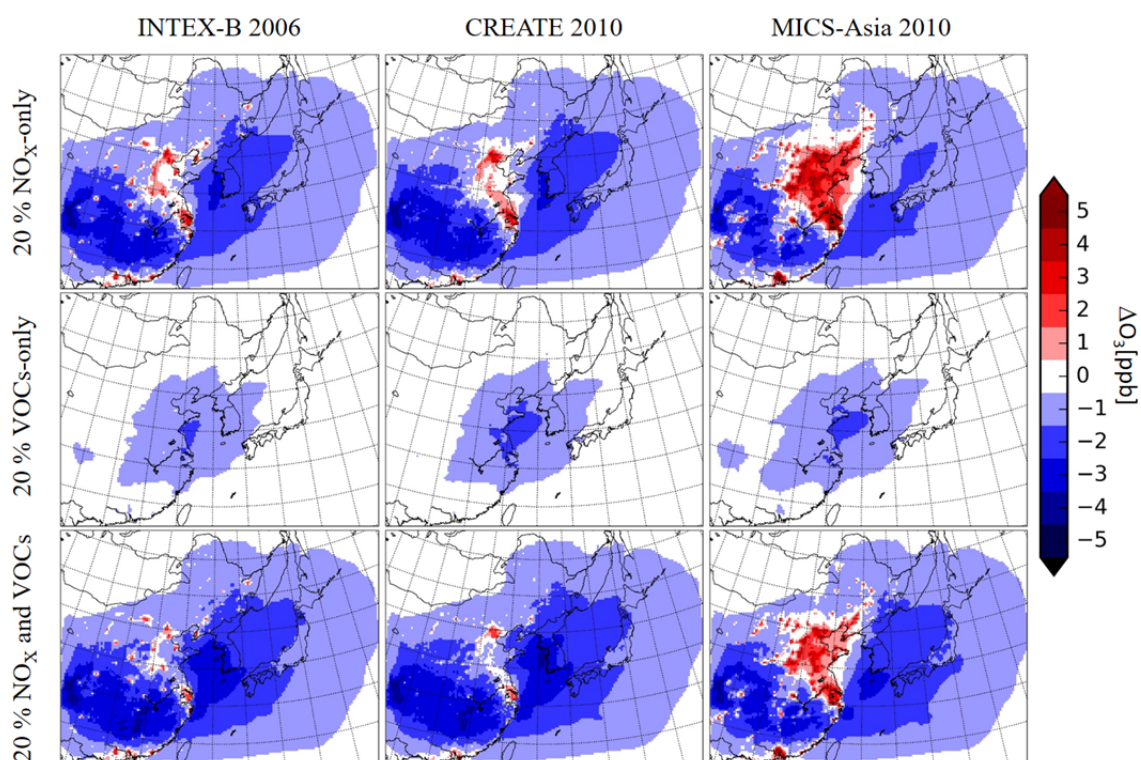
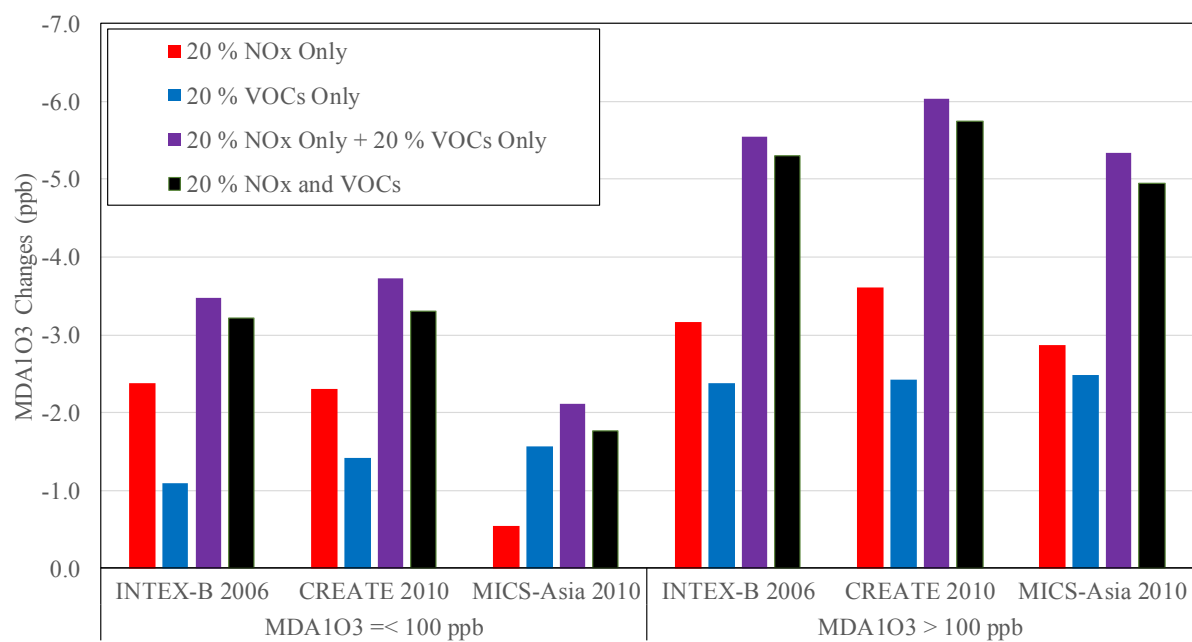
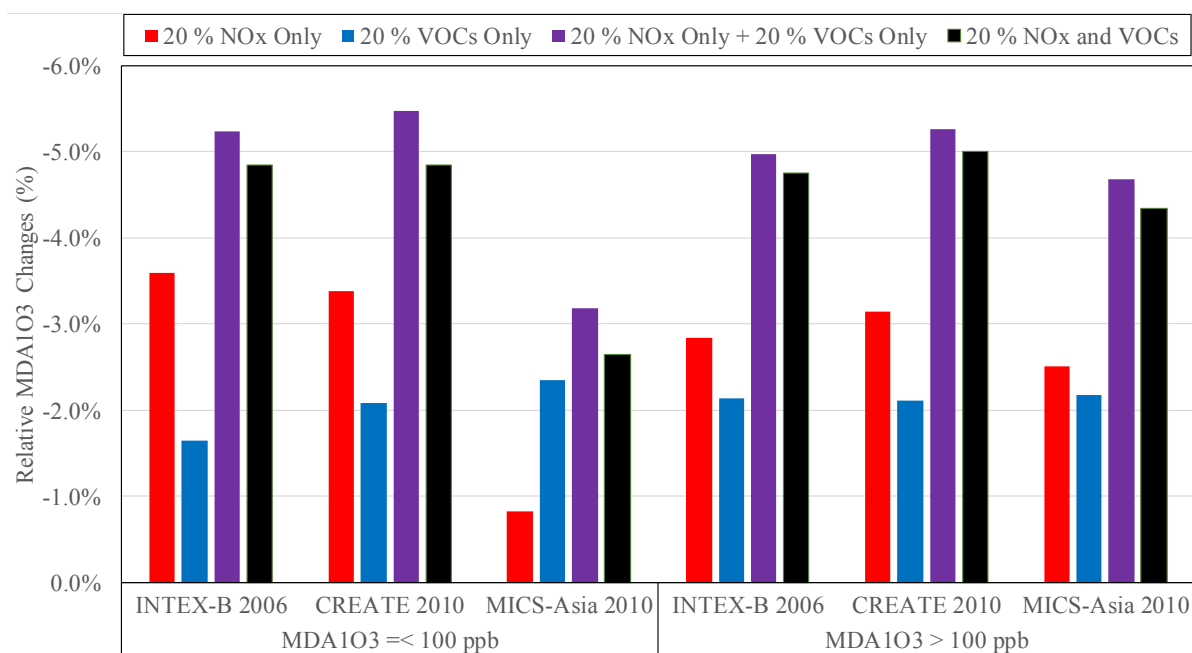


Fig. 10. Episodic average spatial distribution of changes in ozone concentration due to 20% reductions of  $\text{NO}_x$  only (top row), 20% reductions of VOCs only (middle row), and 20% reductions of both  $\text{NO}_x$  and VOCs (bottom row) in Chinese emissions using INTEX-B 2006 (left column), CREATE 2010 (middle column), and MICS-Asia 2010 (right column).

**Table 7.** Spatial averages of modeled MDA1O3 values, by date, that surpass South Korea's one-hour ozone Air Quality Standard in the SMA with the three foreign anthropogenic emissions inventories.

| Date       | INTEX-B 2006 | CREATE 2010 | MICS-Asia 2010 |
|------------|--------------|-------------|----------------|
| 2014-05-23 | 112          | 117         | 116            |
| 2014-05-28 | 102          | 103         | 101            |
| 2014-05-30 | 102          | 106         | 102            |
| 2014-05-31 | 129          | 133         | 138            |

**Fig. 11.** Average changes in ozone due to 20% emissions reductions for days showing MDA1O3 equal to or less than (left) and exceeding (right) the administrative standard, for reductions in only NO<sub>x</sub>, reductions in only VOCs, the sum of reductions in only NO<sub>x</sub> and only VOCs, and reductions in both NO<sub>x</sub> and VOCs at once.**Fig. 12.** Average changes in ozone relative to average MDA1O3 due to 20% reductions in emissions for days showing MDA1O3 equal to or less than (left) and exceeding (right) the administrative standard, for reductions in only NO<sub>x</sub>, reductions in only VOCs, the sum of reductions in only NO<sub>x</sub> and only VOCs, and reductions in both NO<sub>x</sub> and VOCs at once.

defined-ratio reduction cases shown in Figs. 11 and 12. Using the defined-tonnage emissions reduction described in Zhao *et al.* (2013) as an example, the estimated NO<sub>x</sub> reduction from large point sources in China from 2010 to 2015 was about 7,138 TPD, corresponding to 14%, 15%, and 11% of total NO<sub>x</sub> emissions in the INTEX-B 2006, CREATE 2010, and MICS-Asia 2010 inventories, respectively. We recognize that 7,138 TPD is based on a 10% reduction scenario from the emissions inventory used by Zhao *et al.* (2013), which is close to the MICS-Asia 2010 inventory used here in terms of total NO<sub>x</sub> emissions for China.

Table 8 summarizes the absolute and relative (compared to the average) changes in MDA1O3 by exceedance status and emissions inventory. Compared with Figs. 11 and 12, CREATE 2010 exhibits the largest changes in MDA1O3, with INTEX-B 2006 having similar values. MICS-Asia 2010, meanwhile, shows the smallest responses, especially when average MDA1O3 is no greater than 100 ppb. Speculatively, this is because MICS-Asia 2010 has much larger NO<sub>x</sub> emissions in the system overall, which leads to more NO<sub>x</sub> titration conditions across the simulation. When testing the 20% reduction only in NO<sub>x</sub> emissions, MICS-Asia 2010 estimates a change in MDA1O3 of −2.9 ppb for exceeding days. For the same condition, INTEX-B 2006 and CREATE 2010 estimate changes of −3.2 ppb and −3.6 ppb, respectively. Therefore, differences in MDA1O3 changes among all three inventories are within 0.3–0.7 ppb of each other. Meanwhile, a 7,138 TPD NO<sub>x</sub> reduction in China will lead to 1.50–1.78 ppb differences among all three inventories. Thus, when testing future scenario cases including emissions reductions in China, South Korean air-quality planners must, when using predicted ozone air quality with multiple foreign emissions inventories, consider relative reductions in emissions compared to total emissions as well as emissions reductions by defined tonnage.

## CONCLUSIONS

In South Korea, anthropogenic emissions from foreign locations may significantly influence surface ozone concentrations. Therefore, South Korean air-quality managers may need to consider the influence of foreign emissions on South Korean ozone when they develop domestic air-quality management plans. Planners currently favor no specific foreign anthropogenic emissions inventory among the three most commonly used in South Korea: INTEX-B 2006, CREATE 2010, and MICS-Asia 2010. Thus, we examined the characteristics of these foreign emissions inventories by assessing the impacts of inventory choice on modeled ozone concentrations in the SMA. Because of the high ozone concentrations observed in 2014, we chose May

2014 for our study period. Our ozone simulations with all three foreign emissions inventories had comparable results, even though each inventory has specific characteristics in terms of the spatial distribution of emissions. For example, INTEX-B 2006 has much larger NO<sub>x</sub> emissions in large cities in China than the other two inventories. By contrast, the NO<sub>x</sub> emissions of MICS-Asia 2010 are much larger than those of INTEX-B 2006 (and possibly also CREATE 2010) in many other areas, typically small and medium-sized cities and/or locations containing industrial complexes. However, this spatial variability did not seem to affect modeled ozone changes in the SMA in response to test cases of emissions reduction.

For the cases of 20% emissions reduction, we noted that reductions in NO<sub>x</sub> seem to be more effective than reductions in VOCs when choosing the INTEX-B 2006 and CREATE 2010 inventories. Overall, the simultaneous reduction of NO<sub>x</sub> and VOCs resulted in slightly less ozone reduction than the sum of reductions in the individual precursors. We also noted that ozone reductions relative to the predicted ozone concentrations gave similar modeled responses for all three emission inventories on high (> 100 ppb) MDA1O3 days. Finally, using INTEX-B 2006 and CREATE 2010, we showed that defined tonnage reductions lead to similar ozone reduction responses between inventories. As shown in Fig. 10, different synoptic weather patterns could bring the line of transition nearer to or farther from the SMA, with modeled results in this regard depending on the choice of foreign emissions inventory. Further studies should thus be conducted under other meteorological conditions, as there is an implication that the estimated decrease in ozone concentration in the SMA for May 2014 with one foreign emissions inventory may be much more susceptible to meteorological variability than with the other inventories.

We conclude that, when testing future scenario cases including emissions reductions in China, South Korean air-quality planners must, when using predicted ozone air quality with multiple foreign emissions inventories, consider relative reductions in emissions compared to total emissions as well as emissions reductions by defined tonnage. In addition, we recommend using the model's response in a relative sense to minimize differences in modeled ozone reductions due to a choice of foreign emissions inventory, especially for high ozone days. We recognize that our study covers only one month, May 2014, even though South Korea experienced many high ozone days throughout 2014. Because all emissions inventories have different temporal profiles for individual sectors, further studies are warranted to address the effects of these temporal variations on modeled changes in ozone concentration in the SMA in response to reductions in precursor emissions.

**Table 8.** Absolute and relative changes in averaged MDA1O3 for a 7,138 TPD NO<sub>x</sub> reduction in China.

| Emission Inventory                          |                  | INTEX-B 2006 | CREATE 2010 | MICS-Asia 2010 |
|---------------------------------------------|------------------|--------------|-------------|----------------|
| MDA1O3 Change (ppb)                         | MDA1O3 ≤ 100 ppb | −1.58        | −1.86       | −0.08          |
|                                             | MDA1O3 > 100 ppb | −2.12        | −2.96       | −1.22          |
| Relative Response to the Average MDA1O3 (%) | MDA1O3 ≤ 100 ppb | −2.4%        | −2.7%       | −0.1%          |
|                                             | MDA1O3 > 100 ppb | −1.9%        | −2.6%       | −1.1%          |

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## DISCLAIMER

The scientific results and conclusions presented here, as well as any views or opinions expressed herein, are those of the author(s) and do not necessarily reflect the views of the National Oceanic and Atmospheric Administration of the U.S. Department of Commerce.

## SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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