Effects of Vertical Eddy Diffusivities on Regional $PM_{2.5}$ and O_3 Source Contributions

1. Introduction

An effective air pollution reduction plan requires understanding of source–receptor relationships. However, identifying a source–receptor relationship is challenging due to the borderless nature of air pollution. Extent of areas affected by air pollution is changing continuously over time at multiple spatial scales because distribution of air pollutant concentrations is controlled by various physical and chemical processes (Finlayson-Pitts and Pitts Jr, 1999; NARSTO, 2004; Fountoukis and Nenes, 2007; Seinfeld and Pandis, 2016). Moreover, transport of air pollutants often results in transboundary air pollution problems across multiple jurisdictions (Arutyunyan and Aloyan, 1997; Farrell and Keating, 1998; World Health Organization, 2008; Dentener et al., 2010; Rao et al., 2011; Bessagnet et al., 2016; US EPA, 2017). Thus, a source-receptor relationship becomes critical to resolving culpability for air pollution.

Photochemical grid models (PGMs) simulate physical and chemical processes in the atmosphere with first principles as well as parameterizations (Russell and Dennis, 2000; Jacobson, 2005). PGMs can quantify source–receptor relationships explicitly either by tracking air pollutants from sources to receptors or by quantifying sensitivity of air quality at receptors to emission changes at sources (Clappier et al., 2017). Therefore, PGMs have been widely used for assessing upwind source contributions to air quality in downwind areas (Dunker et al., 2002; Yarwood et al., 2007; Kwok et al., 2013, 2015; Clappier et al., 2017).

Among physical processes that control the distribution of air pollutants, vertical turbulent diffusion is a process influencing vertical mixing (Seaman, 2000). Vertical turbulent diffusion results from random circular motions of air parcels (i.e. eddy) with various sizes along the vertical axis (i.e. perpendicular to the ground toward the center of the earth). Vertical turbulent (or eddy) diffusion is an analogous concept derived by applying the concept of molecular diffusion to the behavior of eddies due to their common natures, i.e., randomness. Vertical turbulent diffusion is strong within the planetary boundary layer where we breath air, especially in the daytime, when the thermal buoyant force is intense (Stull, 1988). Vertical turbulent diffusivity (Kv) is a model parameter employed in PGMs to simulate the strength of vertical turbulent diffusion following the concept of molecular diffusivity. Kv has been recognized as a significant model parameter controlling vertical mixing of air pollutants from the dawn of modern PGMs (Scheffe and Morris, 1993; Nowacki et al., 1996).

Past studies identified that Kv in PGMs significantly influences the overall modeling accuracy by controlling how models distribute pollutants vertically near sources and, eventually, how far pollutants can be transported (Liu and Westphal, 2001; Liu et al., 2003; Byun et al., 2007; Queen and Zhang, 2008; Han et al., 2009; Misenis and Zhang, 2010; Tang et al., 2011; Castellanos et al., 2011; Fountoukis et al., 2013). However, previous studies almost exclusively examined effects of Kv on simulated bulk air pollutant concentrations without taking into account the effects of Kv on source contributions. In this study, we examined the effects of Kv on not only the simulated bulk air pollutant concentration but also the modeled source contribution estimates.

2. Method

2.1. Modeling Domain, Representative Episode Selection, and Model Performance Evaluation

Our primary goal was to illustrate the effects of Kv on source contribution estimates as well as modeled bulk air pollutant concentrations. To accomplish our study goal, a modeling domain needs to show strong transboundary air pollution that results in a challenging design question for effective local control strategies at downwind areas considering upwind influences. In addition, to assess the effects of Kv on modeled source-receptor relationship properly, we needed observations at ground level as well as aloft to evaluate model performance rigorously. Therefore, for this study, we simulated air quality in the Northeast Area region because recent studies indicated that air quality in South Korea is heavily affected by domestic and regional foreign sources (Oh et al., 2010; Lee et al., 2013; Oh et al., 2015; Shin et al., 2016; Kim et al., 2017). Also, in May and June of 2016, a field campaign named Korea–United States Air Quality Study (KORUS-AQ; https://espo.nasa.gov/korus-aq/content/KORUS-AQ) was conducted as an international collaboration between National Institute of Environmental Research of South Korea and National Oceanic and Atmospheric Administration of USA. During the KORUS-AQ campaign, extensive observations were made aloft with aircrafts for O_3, $PM_{2.5}$, and their precursors. The modeling domains are shown in Figure 1. We focused on the Seoul Metropolitan Area (SMA, Figure 2) since it is a critical area in the context of South Korea's air quality management because it includes the capital of South Korea, Seoul, and has a population of about > 25 M people as of 2018 (Statistics Korea, 2019).

During the May 2016, nine days were over the daily $PM_{2.5}$ National Ambient Air Quality Standard (35 $\mu g/m^3$) of South Korea and all nine days were between May 15 and May 31, 2016. Thus, we chose May 15–31, 2016, as the modeling period with a 10-day ramp-up period starting from May 6, 2016 (Figure 3). KORUS-AQ Rapid Science Synthesis Report (2017) noted that air quality in SMA for May 17–22 was heavily influenced by the local sources while air quality in SMA for May 25–28 was dominated by transboundary transport. During these two periods, two major inorganic $PM_{2.5}$ constituents (i.e. sulfate and nitrate) showed similar relative contributions to the total $PM_{2.5}$ concentration for each period. Because we attempted to examine source contributions in 3D space, model performance evaluation at high altitudes was crucial. During the modeling period, DC-8 aircraft measurements made as part of the KORUS-AQ are available for eight days (Figure 3). We provide details about our approach for model performance evaluation in the "Approach to Conduct Model Performance Evaluation" section of the Supplementary Material.

After considering three factors - (1) days exceeding South Korea's daily $PM_{2.5}$ NAAQS (May 17 and 25-28), (2) the periods heavily influenced by the local sources (May 17–22) and dominated transboundary transport (May 25–28), and (3) the availability of DC-8 aircraft measurements (May 17, 18, 20, 22, 25, 26, 30, and 31) - we selected May 17, 25, and 26, 2016 for detailed analysis initially (Figure 3). However, our examination of time series for aircraft observation and modeled concentrations aloft showed significant caveats on May 25, 2016 (Figure S8). Specifically, significant underestimation around 1,000 m altitude (10:30 AM-12:00 PM on May 15 KST) and gross overestimation near ground (12:00 PM-1:00 PM on May 15 KST) exhibited great potential to introduce model bias in source contribution analysis results. Therefore, we limited our detailed analysis to two high $PM_{2.5}$ days with sufficient model performance because vertical contribution analyses require solid model performance evaluation with ample observations at ground level as well as aloft. These two days that we selected for detailed analyses should represent days of heavy local source influence and those of transboundary transport domination appropriately and sufficiently.

2.2. Model configurations

For meteorological modeling, we used the Weather Research and Forecasting (WRF) model (Skamarock and Klemp, 2008) v3.4.1 with National Centers for Environmental Prediction Final Analysis (NCEP-FNL) products for initial and boundary conditions. The WRF configuration used for this study is summarized in Table 1. Land cover data used in WRF modeling are displayed in Figure S1. Emissions inventories used were the Comprehensive Regional Emissions Inventory for Atmospheric Transport Experiment (CREATE) 2015 for foreign anthropogenic emissions (Jang et al., 2019) and the Clean Air Policy Support System (CAPSS) 2016 for domestic anthropogenic emissions, which is an updated emission inventory based on previously reported data (Lee et al., 2011). For biogenic emissions, we utilized the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 for biogenic emissions (Guenther, 2006). We processed emission inventory data with Sparse Matrix Operator Kernel Emissions (SMOKE) for modeling-ready emissions input files.

For air quality modeling, we chose the Comprehensive Air Quality Model with eXtension (CAMx) version 6.2 (Ramboll-Environ, 2016). We ran CAMx in the two-way nested modeling mode. In this mode, CAMx calculates pollutant concentrations in a 27-km domain for each numerical integration time step first and uses its results as initial and boundary conditions for modeling on the 9-km domain internally. Then, CAMx aggregates pollutant concentrations in 9-km cells to calculate concentrations in the corresponding grid cells on the 27-km domain. In other words, the estimated pollutant concentrations in a grid cell on the 27-km modeling domain inside the 9-km modeling domain is a spatial average of modeled pollutant concentrations in 9 grid cells on the 9-km modeling domain within the 27-km grid cell. To prepare meteorological inputs for CAMx, we used the WRFCAMx preprocessor, wherein a minimum Kv value can be set.

Specifically, we studied the effects of minimum Kv values that are set for the bottom few modeling vertical layers representing the strength of vertical turbulence near ground, where many sources and receptors are located. For our study, we set the minimum Kv value at 1.0 m^2/s over all modeling grid cells for the base modeling case ("Run A") to maintain consistency with our previous modeling studies (Kim et al., 2017a; Kim et al., 2017b). For a sensitivity modeling case ("Run B"), we set the minimum Kv value in two steps: (1) setting the minimum Kv value as 0.1 m^2/s because past studies in the US and Europe reported that Kv value of 1.0 m^2/s may result in excessive vertical mixing for certain conditions such as the nighttime or rural areas (ENVIRON, 2011; O. US EPA, 2017; Oikonomakis et al., 2018); and (2) using KVPATCH v6 to set an alternative minimum Kv value for areas classified as urban at 1.0 m^2/s at the 1st layer and adjusted Kv values within 200 m from the ground to account for vertical mixing characteristics such as heat island effects (Ramboll, 2014) in urban areas. The urban classification is based on the land use/landcover data used in the WRF. Thus, our Kv value adjustments inherit uncertainties in landuse/landcover from the WRF dataset (Figure S1). For the modeling in this study, the heights at the top of 1st, 2nd, 3rd, and 4th layers were approximately 30, 80, 160, and 250 meters above the ground level. Photolysis rate input files were prepared with the Tropospheric Visible and Ultraviolet and O_3MAP utilities to account for effects of stratospheric O_3 on available actinic flux in the troposphere. The CAMx configuration used for this study is summarized in Table 2.

For source apportionment, we used the Particulate Source Apportionment Technology (PSAT) instrumented in CAMx (Yarwood et al., 2007; Wagstrom et al., 2008; Ramboll-Environ, 2016). 10 source regions were defined in the 27-km modeling domain as shown in Figure 1. Among 10 regions, 9 regions were defined for specific areas (see the caption of Figure 1 for details). We also stratified boundary conditions (i.e., North, South, East, and West boundary conditions). To examine contribution changes at different model vertical layers due to the vertical diffusivity, we placed PSAT receptors not only at the surface (i.e., the 1st modeling layer) but also aloft over the SMA. Because changes in the surface/low layer mixing at sources will affect the initial mixing of emitted pollutants, we expected that two model runs will show differences in the extent of pollutant transport downwind and the degree of local mixing near receptors. It is especially important when secondary pollutants can be formed by multiple precursors originating upwind and downwind areas; e.g., ammonium sulfate and ammonium nitrate formations during the 2014 high $PM_{2.5}$ event in the SMA when sulfuric and nitric acids originating from China were neutralized with South Korean ammonia (B.-U. Kim et al., 2017).

3. Results and Discussion

3.1. Model performance at ground level

3.1.1. Meteorological model performance

In the 27-km domain, a range of mean biases is from -1.9 °C (NRB) to 0.15 °C (COT) for temperatures, and mean biases range from -0.7 m/s (BTH) to -0.1 m/s (JPN) for wind speeds (Figure S2). In the 9-km domain, a range of mean biases is from -2.0 °C (Sudo) to -0.9 °C (Jeju) for temperatures; and, mean biases range from 0.03 m/s (Sudo) to 1.5 m/s (Jeju) for wind speeds (Figure S3). These low biases should not influence the modeled concentrations in the SMA because modeled winds were generally weak on May 17, 2016, and a distance between the SMA and Jeju was over 450 km. In addition, Gangwon is located downwind of SMA. A more comprehensive performance statistics summary is presented in Supplementary Information (Tables S1 and S2) including North Korean SHMA sites in the 27-km domain as NKR.

3.1.2. Air quality model performance at ground level

Over China, in BTH, PRD, and YRD regions, models underestimated O_3 concentrations significantly overall (Figure S4). Run A showed higher nighttime O_3 concentrations than Run B, which led to high biases during the nighttime in some regions such as the NRB. For $PM_{2.5}$, Run B showed slightly higher biases than Run A during the nighttime in general. This indicates that nighttime $PM_{2.5}$ overprediction was likely linked to primary $PM_{2.5}$ emissions. Apparently, in the RUM region, there was a very large $PM_{2.5}$ event that both Run A and Run B missed around the midnight of May 23. It might be due to an unidentified fire event, but it was concluded that it did not affect the overall model performance, especially on May 26, 2016, when transport was important in the SMA. Performance statistics at Chinese NAAQMN sites are listed in Tables S3 and S4. Overall, the fraction errors were 13%-69% for O_3 and from 24%-83% for $PM_{2.5}$ in China.

Both Run A and Run B at US DOS $PM_{2.5}$ sites showed that the overall $PM_{2.5}$ fraction errors were 8%-96% (Table S5 and Figure S5). In general, nighttime overestimation by Run A and Run B was consistent with what we observed at Chinese NAAQMN sites. Run B showed more nighttime biases and the bias trend seemed to follow the inverse of diurnal boundary layer height changes. Because US DOS $PM_{2.5}$ sites are likely located at urban areas, higher biases in Chinese PSAT regions were likely due to primary $PM_{2.5}$ emissions from rural areas in both models. In the future, primary $PM_{2.5}$ emissions needed to be examined further to reduce overestimation. At the US DOS $PM_{2.5}$ sites in Beijing, we observed > 250 $\mu g/m^3$ at 7 PM on May 26, 2016. However, we concluded that this was not significant because it only lasted an hour.

At South Korean AMS sites (i.e. in the 9-km domain), Run A and Run B showed the fraction errors ranging 11%-21% for O_3 and 39%-60% for $PM_{2.5}$ in South Korea (Tables S6-S7 and Figure S6). Models overestimated O_3 concentrations during the nighttime while they underestimate O_3 concentrations during the daytime in Sudo and Jungboo. This can result from too weak nighttime O_3 removal and too much daytime NO_X titration. In general, Run A showed higher nighttime O_3 concentrations. For total $PM_{2.5}$ mass concentrations, Run B showed slightly higher biases than Run A in general. Similar to what we observed at monitors in China, what was observed at South Korean AMS sites indicates that nighttime $PM_{2.5}$ overprediction is also likely linked to primary $PM_{2.5}$ emissions. However, both models also showed underpredictions for certain days, which may indicate insufficient local emissions or contributions from other regions. Further studies are warranted.

To examine the chemical characteristics of the $PM_{2.5}$ model performance, we compared observed and modeled total and speciated $PM_{2.5}$ mass concentrations focusing the model performance at the Bulgwang Supersite (Figure S7), which is located in the focus area, SMA, even though we examined model performance at all Supersites (not shown here). Both models showed fractional biases ranging from -25.92% ($PM_{2.5}$ by Run A) to 120.89% (crustal by Run B) at the Bulgwang Supersite. More detailed performance statistics for total $PM_{2.5}$, sulfate, nitrate, ammonium, organic matter, elemental carbon, and crustal are presented in Table S8).

3.2. Model performance aloft

Because objectives of this study include the effects of vertical mixing at low modeling layers on the regional transport of air pollutants, it is important to evaluate the model performance aloft. Thus, we conducted model performance evaluation on May 17 and 26, 2016, with aircraft measurements after considering aircraft data availability and significance of high $PM_{2.5}$ events. For both days, the aircraft observed key meteorological variables (e.g. winds and temperatures) and air pollutant concentrations (e.g. sulfate and nitrate). We provide details about DC-8 flight paths on May 17 and 26, 2016, including flight paths with aircraft speeds/directions, ambient temperature, measured wind speeds/directions, and O_3 concentrations on May 17 and 26, 2016 in Figure S9. Flight paths with measured wind speeds/directions clearly showed the influence of foreign transport aloft on May 17 and from ground to aloft on May 26.

Figure S10 shows a comparison between the observed and modeled meteorological variables on May 17 and 26, 2016, along the DC-8 flight paths. For the flight duration on May 17, 2016, the model showed consistency with observations for meteorological variables. The exceptions were wind speeds at low altitudes (< 1,000 m). We also observed that overestimation of wind speeds at ground monitors. During the flight on May 26, 2016, the model showed good agreement with observations for meteorological variables including wind speeds at low altitudes (< 1,000 m). Model performance evaluation with aircraft measurements provided good confidence with meteorological fields used for this study in general. In particular, the model performance on May 26, 2016, was important because this day was examined in detail for non-local contributions.

Based on a previous report (KORUS-AQ Rapid Science Synthesis Report, 2017) and DC-8 flight paths (Figure S11), we considered May 17, 2016, as a local influence day. Note that "local influence" is defined as a relative term. That is, "local influence" days are when domestic contributions are relatively higher than other days because foreign contributions can be over 50% most days. For May 17, 2016, we focused our performance evaluation more for areas near Seoul. For O_3 and its precursors, Run A and Run B showed very similar results (Figure S11). Overall, model performance for sulfate was good while both models underestimated organic carbon significantly. The overall model biases in May 17 for sulfate and nitrate were -0.16 $\mu g/m^3$ and -0.62 $\mu g/m^3$, respectively. Based on a previous report (KORUS-AQ Rapid Science Synthesis Report, 2017) and DC-8 flight paths (Figure S12), we considered May 26, 2016, as a transboundary transport day. Overall, we concluded that the overall model performances were sufficient for this study. The overall model biases in May 26 for sulfate and nitrate were 4.1 $\mu g/m^3$ and -2.4 $\mu g/m^3$, respectively. For more detailed discussion, we present "Detailed model performance analysis with DC-8 aircraft measurement" in the Supplemental Material.

3.3. Sensitivity of modeled concentration distributions to minimum Kv values

An earlier study (KORUS-AQ Rapid Science Synthesis Report, 2017) reported that $PM_{2.5}$ concentrations in South Korea on May 17, 2016, were likely influenced by domestic emissions. Figure 4 shows the spatial distribution of daily average modeled $PM_{2.5}$ and SIA concentrations in the 9-km modeling domain on May 17, 2016. It also displays impacts of changing a minimum Kv value used in modeling the spatial distribution of $PM_{2.5}$ and SIA concentrations. Compared with Run A, $PM_{2.5}$ concentrations in Run B were much higher (> 10 $\mu g/m^3$) around the west coast of South Korea in general. The domain-wide maximum difference was 36.0 $\mu g/m^3$ in the vicinity of the west cost of the Choongnam province located at the southwest of SMA. In the SMA area, Run B resulted in 5.0–13.2 $\mu g/m^3$ higher $PM_{2.5}$ concentrations in the 9-km grid cells than Run A.

According to a previous work (KORUS-AQ Rapid Science Synthesis Report, 2017), May 26, 2016, is considered as a day on which transboundary transport influenced $PM_{2.5}$ concentrations in South Korea. Figure 5 shows the spatial distribution of daily average modeled $PM_{2.5}$ and SIA concentrations in the 27-km modeling domain on May 26, 2016. With weaker minimum vertical mixing at lower layers in Run B, total $PM_{2.5}$ concentrations in the many parts of inland China were predicted to be higher according to Run B than Run A. However, we noted that SIA concentrations were not the sole primary cause. In addition, total $PM_{2.5}$ and SIA concentrations according to Run B were much lower over the most of Yellow Sea. Around several inland areas along the west coastline of South Korea, $PM_{2.5}$ concentrations in Run B were much higher (> 10 μg/m^3) than Run A. $PM_{2.5}$ concentration differences between Run A and Run B ranged from -5 μg/m^3 to 89.1 μg/m^3 in the 27-km modeling grid cells along the west coastline of China and from -14.0 μg/m^3 to 0.6 μg/m^3 in the 9-km modeling grid cells inside the SMA area. High sulfate concentrations (> 20 μg/m^3) in the 27-km domain occurred in the southern BTH region, Yellow Sea, majority of South Korea, and East Sea on May 26, 2016. This highlights that sulfate can be a significant $PM_{2.5}$ constituent during the summertime as well as wintertime, as reported by (B.-U. Kim et al., 2017) for when South Korea experienced high PM events due to transboundary transport. The significance of sulfate may need to be carefully reassessed in the future because several studies reported that SO_2 emission in China has been declining (Li et al., 2018; Kang et al., 2019; Qu et al., 2019). Overall, Run B estimated higher nitrate concentrations but lower sulfate concentrations over the inland areas of South Korea than Run A on May 26, 2016. In addition, we noticed much larger differences for SIA concentrations near the east coast of China and over Yellow Sea on May 26, 2016 than May 17, 2016.

Overall, a weaker low-level vertical mixing condition (i.e., Run B) seemed to lead to higher SIAs near the SMA, when local influences were important while the same condition resulted in a mixed effect, when transport influences were dominant (i.e., lower sulfate and higher nitrate concentrations). This may imply that Chinese sulfuric acid and/or sulfate were formed slower in Run B because of delayed reactions with oxidants and ammonia due to weaker vertical mixing at the surface. For the same reason, Chinese nitric acid and/or nitrate could be formed more locally (i.e., near sources) in Run B.

3.4. Sensitivity of modeled source contributions to minimum Kv values

We found different spatial distributions of $PM_{2.5}$ concentrations due to variation in the minimum Kv values over the modeling domains. For the SMA, we noticed that lower minimum Kv values led to higher $PM_{2.5}$ concentrations on May 17, 2016 (a local influence day). As we noted in "3.2. Model performance aloft" section, "local influence" is a relative term. For some parts of the SMA, Run B resulted in lower $PM_{2.5}$ concentrations on May 26, 2016 (a transboundary transport day). To explain the concentration differences in the SMA with contribution changes, we examined the vertical distribution of domestic/foreign contributions on May 17 and 26, 2016.

3.4.1. May 17, 2016: Local Influence Day

Figures 6-7 show daily average $PM_{2.5}$ concentrations and daily maximum 1-hour O_3 concentrations over the SMA receptor cells as depicted in Figure 2. On May 17, 2016, daily average $PM_{2.5}$ concentrations at the 1st layer estimated by Run A and Run B were 43.1 μg/m^3 and 49.9 μg/m^3, respectively. Domestic contributions estimated by Run A and Run B were 16.7 μg/m^3 and 19.7 μg/m^3, respectively. 43.4% (3.0 μg/m^3) of the modeled $PM_{2.5}$ concentration differences (6.9 μg/m^3) could be explained by modeled domestic contribution changes due to the choice of the minimum Kv value. Relative ratios of foreign contributions to domestic contributions were 1.6 and 1.5 according to Run A and Run B, respectively. This indicates a lower minimum Kv value seems to result in domestic and foreign contribution changes to a similar degree when the local influence is dominant. This also implies that local emission controls for high $PM_{2.5}$ event days can be effective regardless uncertainties in foreign influences due to various reasons.

Daily maximum 1-hour O_3 values in the SMA at 3 PM on May 17, 2016, were similar between two modeling cases (56.1 ppb by Run A and 56.4 ppb by Run B). Foreign contributions estimated by Run B, except that those from BTH, were larger at low altitudes (e.g. the 5th layer or below) while contributions of domestic sources (e.g. 0.2 ppb at the 3rd – 6th layers) and boundary conditions (esp. North BC) estimated by Run B are smaller by than Run A. The North BC contribution was smaller up to the 10th layer (c.a. 1000 m) in Run B than Run A. Domestic contributions were very similar between Run A and Run B at the 1st layer while the domestic contribution according to Run B was smaller from the 2nd layer to 8th layer than that according to Run A. Apparently, YRD's contribution is consistently larger at all layers for Run B than Run A. Excluding BCs, the two major contributors were BTH and South Korea. Their contributions were 10.2 ppb (BTH) and 9.1 ppb (South Korea) according to Run A and 9.9 ppb (BTH) and 9.0 ppb (South Korea) according to Run B. Overall, foreign contributions were 5 times of the domestic contributions according to both Run A and Run B. Boundary conditions accounted for 48% and 47% of the total O_3 concentrations according to Run A and Run B, respectively. This emphasizes the importance of BCs for good O_3 model performance although it may not change the relative contributions of domestic and foreign sources.

May 26, 2016: Transboundary Transport Day

Figures 8 and 9 show daily average $PM_{2.5}$ concentrations and daily maximum 1-hour O_3 concentrations over the SMA receptor cells on May 26, 2016. In general, foreign contributions were smaller according to Run B at low altitudes while domestic contributions were larger at the 1st and 2nd layers. NRB, YRD, and COT showed a non-monotonic pattern of contribution differences for Run A and Run B in the SMA along the altitude. Contributions of NRB, YRD, and COT were smaller according to Run B up to the 6th layer and became larger between the 7th and 10th layers. It implies that a source area specific foreign contribution can be sensitive to model configuration such as Kv. Also, it highlights the importance of understanding how foreign originated air pollutants are transported aloft to better explain foreign contributions at downwind receptors. On May 26, 2016, the estimated daily average $PM_{2.5}$ concentrations at the 1st layer according to Run A and Run B were 72.0 $\mu g/m^3$ and 65.9 $\mu g/m^3$, respectively. Domestic contributions estimated by Run A and Run B were 18.2 $\mu g/m^3$ and 19.1 $\mu g/m^3$ (25.2% and 29.0% of total $PM_{2.5}$ concentrations), respectively. Relative ratios of foreign contributions to domestic contributions were 3.0 and 2.4 according to Run A and Run B, respectively. One interesting observation was that South Korea's domestic contributions according to Run B on May 17 (19.7 $\mu g/m^3$) and May 26 (19.1 $\mu g/m^3$), 2016, showed little differences while those according to run A on May 17 (16.7 $\mu g/m^3$) and May 26 (18.2 $\mu g/m^3$), 2016, showed relatively large differences. This implies that the landuse/landcover specific Kv min value (i.e. Run B; 1.0 m^2/s for urban areas and 0.1 m^2/s for rural areas) leads to more converging domestic contribution estimates than a constantly higher Kv value (i.e. Run A; 1.0 m^2/s) across receptor areas.

Daily maximum 1-hour O_3 concentrations in the SMA at 1 PM on May 26, 2016, were similar for the two modeling cases (78.7 ppb by Run A and 78.0 ppb by Run B). Unlike May 17, 2016, Run A estimated higher (0.7 ppb) daily maximum 1-hour O_3 concentrations in the SMA on May 26, 2016, than Run B. At lower altitudes, YRD was the major contributor. However, at higher altitudes

(> 1000 m), NRB became the major contributor. The apparent O_3 concentration differences at the surface level were due to lower contributions from YRD and West BC according to Run B than Run A. The overall differences were compensated by lower YRD contributions and higher COT and NRB contributions according to Run B than Run A. Domestic contributions were similar for Run A and Run B. Apparently, YRD's contribution was consistently lower at all layers for Run B than Run A. Including BCs, the major contributor was YRD and its contributions were estimated to be 27.2 ppb and 25.3 ppb by Run A and Run B, respectively. The 2nd highest contributor was NRB (16.0 ppb and 16.3 ppb, respectively). Overall, foreign contributions were 11 times of the domestic contributions according to both Run A and Run B. Domestic contributions were 6.4 ppb according to both Run A and Run B. Compared with May 17, 2016, domestic contributions decreased by 29% according to both Run A and Run B on May 26, 2016.

4. Conclusion

In this study, we examined the effects of minimum Kv values on modeled contributions by simulating the air quality in the Northeast Asia region. Because of the availability of various data including aircraft measurements and the strategic importance of air quality management in South Korea, we focused our analysis for the SMA on May 17 (a local influence day) and 26 (a transboundary transport day), 2016. Two model runs were performed with minimum Kv values of 1.0 m2/s (Run A) and 0.1 m2/s with additional adjustment below the altitude of 200 m (Run B).

For May 17, 2016 (a local influence day), Run A and Run B estimated that domestic $PM_{2.5}$ contributions were 16.7 $\mu g/m^3$ and 19.7 $\mu g/m^3$. Relative domestic contributions to total $PM_{2.5}$ concentrations were similar (38.8 % vs 39.4 %) according to the two model runs although modeled total $PM_{2.5}$ mass concentrations in the SMA (43.1 $\mu g/m^3$ and 49.9 $\mu g/m^3$) were quite different as large as 6 $\mu g/m^3$. We found a similar pattern for O_3 on the same day. However, on May 26, 2016 (a foreign influence day), relative ratios of foreign $PM_{2.5}$ contributions to domestic $PM_{2.5}$ contributions were quite different (3.0 by Run A and 2.4 by Run B) according to the two model runs. Overall, the lower minimum Kv value near surface led to higher $PM_{2.5}$ concentrations over the South Korean peninsula. However, this was not necessarily true for the SMA. On May 26, 2016 (a foreign influence day), the lower minimum Kv value resulted in lower $PM_{2.5}$ concentrations in the SMA. It turns out that foreign contributions were much lower on May 26, 2016, with lower minimum Kv values while the domestic contribution became larger. Apparently, a choice of minimum Kv value used in a modeling study caused greater sensitivity on days influenced by the transboundary transport of air pollution.

As we discussed in the previous section, vertical mixing differences can lead to variations in chemical reaction timing in Chinese source areas by influencing initial mixing near sources, then change how primary and secondary pollutants as well as their precursors are transported in terms of distance and altitude. At the same time, we noted that modeled absolute domestic contributions were similar for a local influence day and a transboundary transport day with the lower minimum Kv value. If we can consider this as an evidence for more solid estimation in domestic contributions, we may be able to estimate bias-adjusted foreign contributions in the future. This also implies that it may be desirable for air quality modelers to perform two sets of modeling with higher and lower minimum Kv values when they conduct modeling for developing air quality plans that should consider domestic/foreign contributions.

The limitation of this study was that only two focus days were examined during summertime in 2016 even though those two days might represent sufficiently local against transboundary transport dominant days. Because seasonality and inter-annual variations along with rapid emission changes in the Northeast Asia region can play critical roles, further studies are warrantied to examine the effects of a critical model parameter, minimum Kv, on modeled air pollutant concentrations and estimated source contribution analyses for longer terms, e.g. annual or multi-year simulations.

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