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The Impact of Foreign SO_2 Emissions on Aerosol Direct Radiative Effects in South Korea

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Abstract: This study examined the impact of foreign SO_2 emission changes on the aerosol direct radiative effects (ADRE) in South Korea. Simulations that applied basic emissions (BASE) and simulations that applied reduced SO_2 emissions from foreign sources (R_FSO2) were performed, respectively, using the Weather Research and Forecasting (WRF)-Community Multiscale Air Quality (CMAQ) two-way coupled model. In addition, the difference between the two experimental results was calculated (i.e., R_FSO2 minus BASE) to quantitatively identify the impact of foreign SO_2 emission reduction. The reduction in foreign SO_2 emissions caused a decrease in the concentration of SO_2 flowing in from overseas to South Korea. As a result, a clear decrease in SO_4^{2-} concentration was shown mainly in the southwest coast of South Korea. The difference in $PM_{2.5}$ concentration in South Korea according to the foreign SO_2 emission reduction did not correspond to the difference in SO_4^{2-} concentration; it was determined in a complex way by the changes in SO_4^{2-} concentration caused by SO_2 concentration changes, and the subsequent series of changes in NO_3^- and NH_4^+ concentrations. The differences in SO_4^{2-} and $PM_{2.5}$ concentrations caused by the foreign SO_2 reduction also affected the ADRE changes in South Korea. The distribution of ADRE difference between the two experiments was not consistent with the distribution of $PM_{2.5}$ concentration difference, but it was very similar to the distribution of SO_4^{2-} concentration difference. These results imply that the ADRE of South Korea is not simply proportional to $PM_{2.5}$ concentration and may be determined by concentration changes of SO_4^{2-}.

Keywords: SO_2 emissions; SO_4^{2-}; WRF–CMAQ two-way coupled model; aerosol direct radiative effects

1. Introduction

Sulfate (SO_4^{2-}) is a major component of $PM_{2.5}$, and the generation of high-concentration SO_4^{2-} is closely related to SO_2 emissions [1]. China is one of the world's largest countries emitting SO_2 [2], and the SO_4^{2-} concentration of South Korea located in the downwind region of China is greatly affected by SO_2 transported from China [3–6]. According to Lee et al. [3], 8–12% of SO_2 in Seoul of South Korea originated from China, and Choi et al. [7] reported that the rapid increase in SO_4^{2-} concentration during high-concentration $PM_{2.5}$ episodes in South Korea was due to the transport of foreign SO_2 emissions. Because SO_2 emitted from China produces $PM_{2.5}$ components such as SO_4^{2-} and ammonium sulfate ($(NH_4)_2SO_4$) through chemical reactions in the process of being transported across the Yellow Sea to South Korea [6], it affects the $PM_{2.5}$ concentration changes in South Korea.

$PM_{2.5}$ concentration change in the atmosphere may cause changes in the meteorological factors such as solar radiation and temperature, and these changes affect the planetary boundary layer (PBL) height, photolysis rates, and the like, which again cause changes in $PM_{2.5}$ concentration [8,9]. The scattering and/or reflection of solar radiation by aerosols in the atmosphere, affecting the meteorology and air pollutants' concentration [10–13], is called Aerosol Direct Radiative Effects (ADRE). Studies of ADRE have been carried out in a variety of ways, mainly using a two-way coupled model (e.g., Weather Research and Forecasting (WRF)-Community Multiscale Air Quality (CMAQ), and WRF-Chem) that can take into account the effects of aerosols. For example, Hong et al. (2017) [14] reported an improved accuracy of O_3 and $PM_{2.5}$ simulation results by considering ADRE, and Nguyen et al. (2019) [15] analyzed that the increase in $PM_{2.5}$ concentration attributes to a decrease in meteorological factors due to ADRE, and Jung et al. (2019) [16] confirmed that ADRE increases the concentration of $PM_{2.5}$ constituents such as SO_4^{2-} and NO_3^-. Many other studies have also reported that ADRE affects surface $PM_{2.5}$ concentration, long-range transport, and even human health [17–19]

The ADRE may be large when the concentration of aerosol in the atmosphere is high (i.e., when the concentration of $PM_{2.5}$ is high) [15] but it does not simply increase in proportion to $PM_{2.5}$ concentration. According to Yoo et al. [13], South Korea's ADRE varies depending on the concentration of each component rather than the total concentration of $PM_{2.5}$. Particularly, SO_4^{2-} concentration is reported to play an important role. Therefore, the concentration of SO_2, which is a major precursor of SO_4^{2-}, affects the ADRE, and consequently, SO_2 transported from China can affect the ADRE distribution in South Korea. However, because the previous study only estimated the conclusion qualitatively and did not provide quantitative evidence, additional investigation and analysis are called for more generalization.

This study thus aimed to quantitatively analyze the impact of foreign SO_2 emissions on the $PM_{2.5}$ (especially SO_4^{2-}) concentration and the ADRE distribution in South Korea. To examine the changes in SO_4^{2-} concentration over South Korea based on the foreign SO_2 emission changes, we applied the brute force method (BFM) to this study. BFM is a method of quantitatively analyzing the differences in modeling results due to emission reduction, which is used in various studies related to contribution analysis [20–23]. The WRF–CMAQ two-way coupled model [24], which facilitates the quantitative calculation of the ADRE, was used to analyze the characteristics of changes in the ADRE distribution according to SO_4^{2-} concentration changes. This is intended to conduct a quantitative analysis that was not performed in previous studies, but very important to better understand the effects of ADRE on $PM_{2.5}$ concentration.

2. Method

2.1. Modeling System

In this study, we used the WRF–CMAQ two-way coupled model, which is a combination of the weather model, Weather Research and Forecasting (WRF, ver. 3.8), and the air quality model, Community Multiscale Air Quality (CMAQ, ver. 5.2) for numerical simulation. This model can quantitatively analyze the differences in $PM_{2.5}$ concentration and the Aerosol Direct Radiative Effects (ADRE) results according to the emission difference because the direct feedback effect of aerosol can be selectively applied when the numerical simulation is performed. For the modeling domain, Northeast Asia including South Korea was selected. The horizontal resolution was configured to be 12 km (280 × 230) and the total number of vertical layers was 30 (Figure 1). The final operational global analysis data (FNL) of National Centers for Environmental Prediction (NCEP) with the horizontal resolution of 1.0° × 1.0° was used as the initial and boundary data of the WRF model, and the Model Inter-Comparison Study for Asia (MICS-Asia) [2] with the horizontal resolution of 0.25° × 0.25° was used as the anthropogenic emission data. The numerical simulation period was one month in February 2015, and the detailed settings for the modeling were applied in the same way as in Yoo et al. [13].

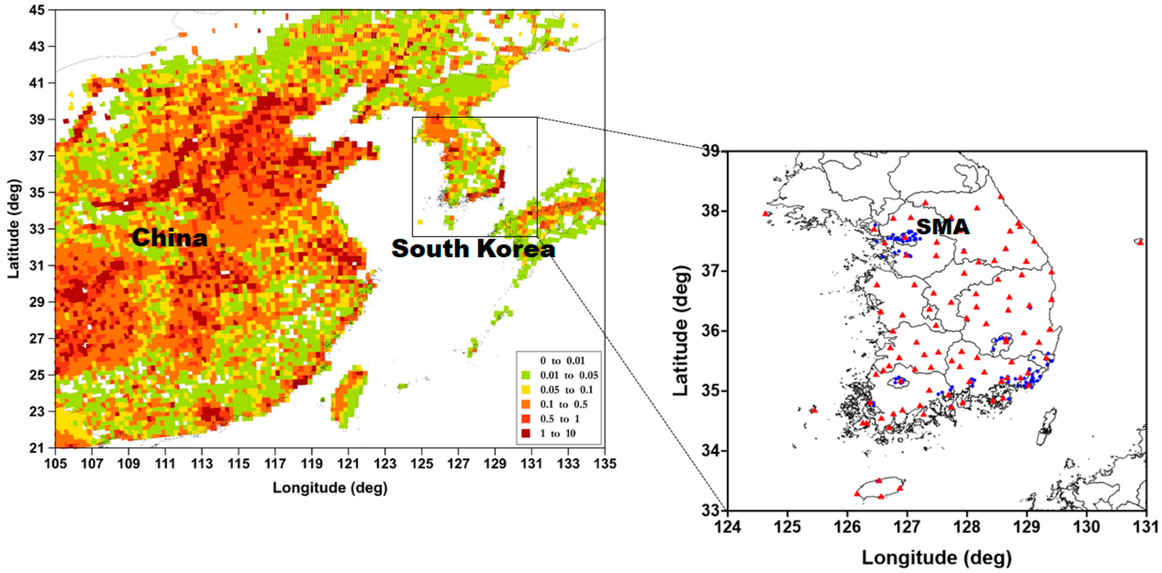


Figure 1. The modeling domain and SO_2 emissions (moles/s) distribution (**left**), and the location map of South Korea (**right**). The right panel shows the locations of meteorological (red triangles) and air quality (blue circles) observation sites. "SMA" denotes Seoul Metropolitan Area.

2.2. Emission Reduction Method

In this study, the impact of foreign SO_2 emissions on $PM_{2.5}$ and the ADRE in South Korea was analyzed using the BFM. BFM is a method of calculating the estimated contribution from the difference of simulated concentrations after changing the emissions of air pollutants to be analyzed; it is mainly used in research to quantitatively analyze the impact of emissions [25]. Simulations that applied basic emissions (BASE) experiments using basic emission data and the R_FSO2 (i.e., reduced foreign SO_2 emissions) experiment using a 50% reduction in the foreign SO_2 emissions were performed, respectively, to analyze the impact of foreign SO_2 emissions on South Korea, and the numerical simulation results were compared.

2.3. Analysis Method of Aerosol Direct Radiative Effects

The WRF–CMAQ two-way coupled model (WRF–CMAQ, hereafter) used in this study was developed to simulate the effects of aerosol for the rapid and accurate radiative transfer model for general circulation models' (RRTMG) short-wave radiation scheme. First, the results of simulation in the CMAQ model were used to calculate the values such as the aerosol's optical characteristics, scattering, and absorption rate, and these values were fed back to the WRF model to calculate the short-wave radiation. The short-wave radiation values thus calculated affect other meteorological factors (e.g., temperature, PBL height), which again cause the changes in aerosol concentration in the atmosphere. In the WRF–CMAQ model, the ADRE values are calculated through the repetition of this feedback process. In this study, the aerosol feedback effects were applied (and not applied) to the BASE and R_FSO2 experiments, respectively, to calculate the ADRE value for each experiment. Then, we focused the analysis on the difference of results between the two experiments.

2.4. Statistical Analysis

Statistical parameters such as the mean bias (MB), the root mean square error (RMSE), correlation coefficient (R), and the index of agreement (IOA) were used to evaluate the meteorological and air quality simulation results. Each parameter is defined as follows:

$$\mathrm{MB} = \frac{\sum_{i=1}^{n}(P_i - O_i)}{n}, \tag{1}$$

$$\mathrm{RMSE} = \sqrt{\frac{\sum_{i=1}^{n}(P_i - O_i)^2}{n}} \tag{2}$$

$$\mathrm{IOA} = 1 - \frac{\sum_{i=1}^{n}(P_i - O_i)^2}{\sum_{i=1}^{n}\left(\left|P_i - \overline{P}\right| + \left|O_i - \overline{O}\right|\right)^2} \tag{3}$$

$$\mathrm{R} = \frac{\sum_{i=1}^{n}\left[\left(O_i - \overline{O}\right) \times \left(P_i - \overline{P}\right)\right]}{\sqrt{\sum_{i=1}^{n}\left(O_i - \overline{O}\right)^2 \times \sum_{i=1}^{n}\left(P_i - \overline{P}\right)^2}} \tag{4}$$

where n is the number of data pairs, and P_i and O_i are the ith WRF and CMAQ simulated and observed values, respectively. $\overline{P}$ and $\overline{O}$ denote the mean WRF and CMAQ simulated and observed values, respectively.

3. Results

3.1. Model Performance

The meteorological and air quality simulation results were evaluated, respectively, to analyze the accuracy of WRF–CMAQ numerical simulation results. First, the hourly observation values and the model values (BASE simulation) were compared for 94 meteorological observation sites in South Korea (Figure 1) during the analysis period to evaluate the numerical simulation accuracy of WRF, a meteorological model (Figure 2). In the case of 2 m temperature (T2), the model value was overestimated by 0.18 °C compared to the observation value, and the R and IOA were 0.89 and 0.91, respectively, satisfying the baseline (Bias ≤ ±0.5 K; IOA ≥ 0.7) provided by Emery et al. [26]. In the case of 10 m wind speed (WS10), the RMSE was 2.17 m s^{-1}, which was slightly overestimated compared to the baseline (≤2.0 m s^{-1}), but the IOA was 0.75, which satisfied the baseline (≥0.6). The hourly $PM_{2.5}$ observation values and model values (BASE experiment) of 90 observation sites (Figure 1) in South Korea during the analysis period were compared to evaluate the simulation results of CMAQ, an air quality model. The CMAQ simulation results were overestimated compared to the observation values, but the IOA value was 0.77, which was high, indicating that the temporal variation of observation values was well simulated overall.

3.2. SO_4^{2-} and $PM_{2.5}$ Concentration Changes in South Korea due to Reduction of Foreign SO_2 Emissions

In this study, we examined the impact of foreign SO_2 emissions on the SO_4^{2-} and $PM_{2.5}$ concentration changes in South Korea. Since air pollutants are mainly distributed below the PBL height, the mean values within the PBL height were used when calculating the difference of results between the experiments (R_FSO2–BASE). As illustrated in Figure 3a, the SO_4^{2-} concentration difference between the two experiments showed a negative value in the whole domain and a decrease of 2.66 μg m^{-3} on average. This is the result of the decrease in SO_2 (main precursor of SO_4^{2-}) transport from upwind regions (e.g., China) to South Korea due to the reduced foreign SO_2 emissions. SO_2 oxidation reactions (R1 to R3) decrease due to the decline of SO_2 concentration in the atmosphere. As a result, the production of H_2SO_4, a direct precursor of SO_4^{2-} slowed down, which in turn led to the decrease in SO_4^{2-} concentration. The distribution of SO_4^{2-} concentration difference was mainly on the southwest coast and the east coast of South Korea. According to the analysis, this was because SO_2 transported from overseas during the case period (February 2015) was mainly on the southwest coast (SO_2 transported from eastern China by southwestern wind) and the east coast (SO_2 transported from Liaodong and North Korea by northwestern wind):

$$SO_2 + OH \rightarrow HOSO_2 \tag{R1}$$

$$HOSO_2 + O_2 \rightarrow HO_2 + SO_3 \tag{R2}$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad \text{(R3)}$$

$$2NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4 \quad \text{(R4)}$$

$$NH_3 + HNO_3 \rightarrow NH_4NO_3 \quad \text{(R5)}$$

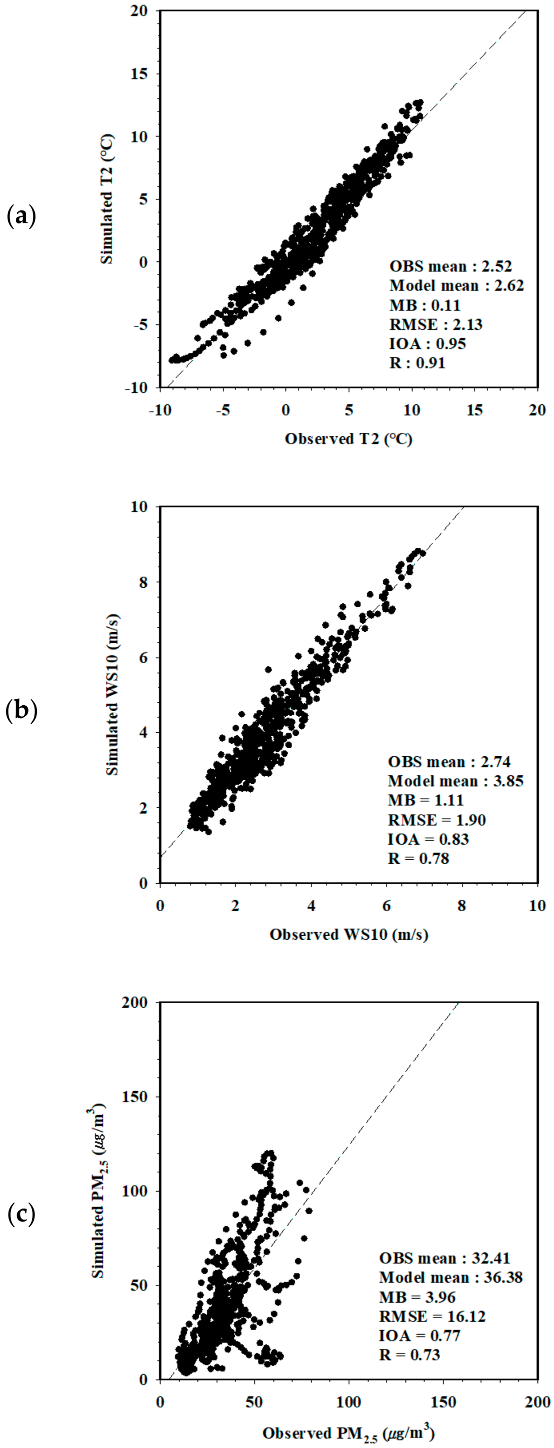


Figure 2. The comparison of hourly observation and model values for (**a**) 2 m temperature (T2), (**b**) 10 m wind speed (WS10), and (**c**) $PM_{2.5}$ concentration. "OBS" is observation, "MB" is mean bias, "RMSE" is root mean squared error, "IOA" is index of agreement, and "R" is correlation coefficient.

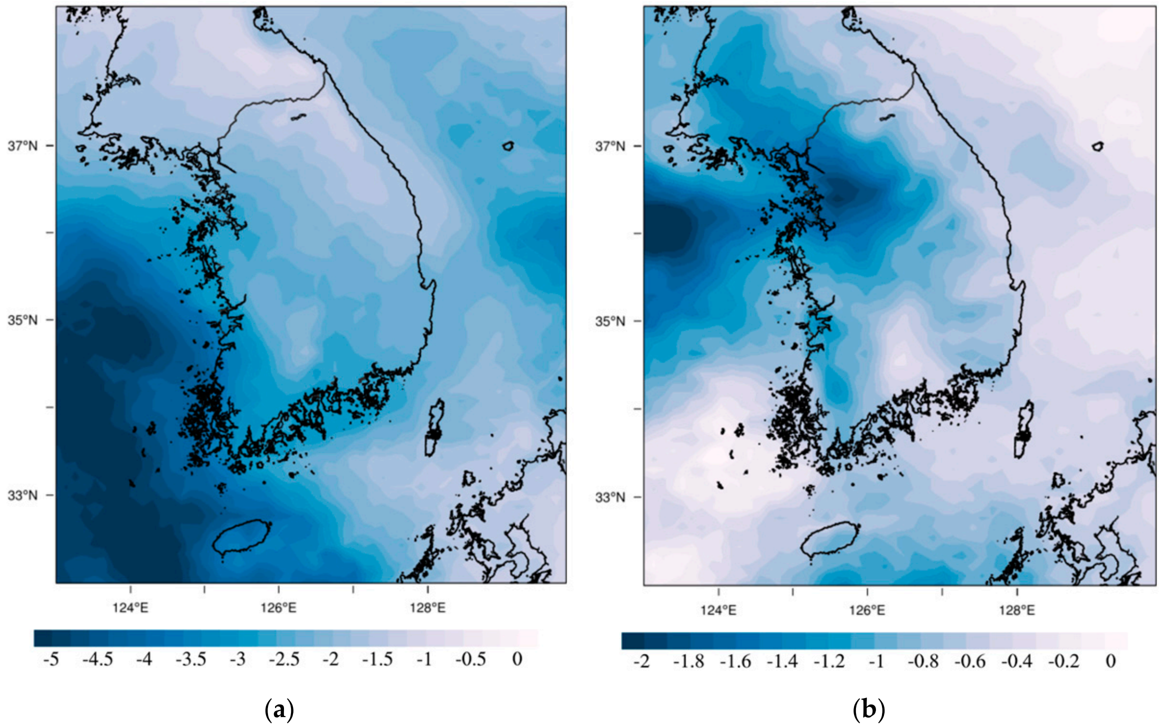
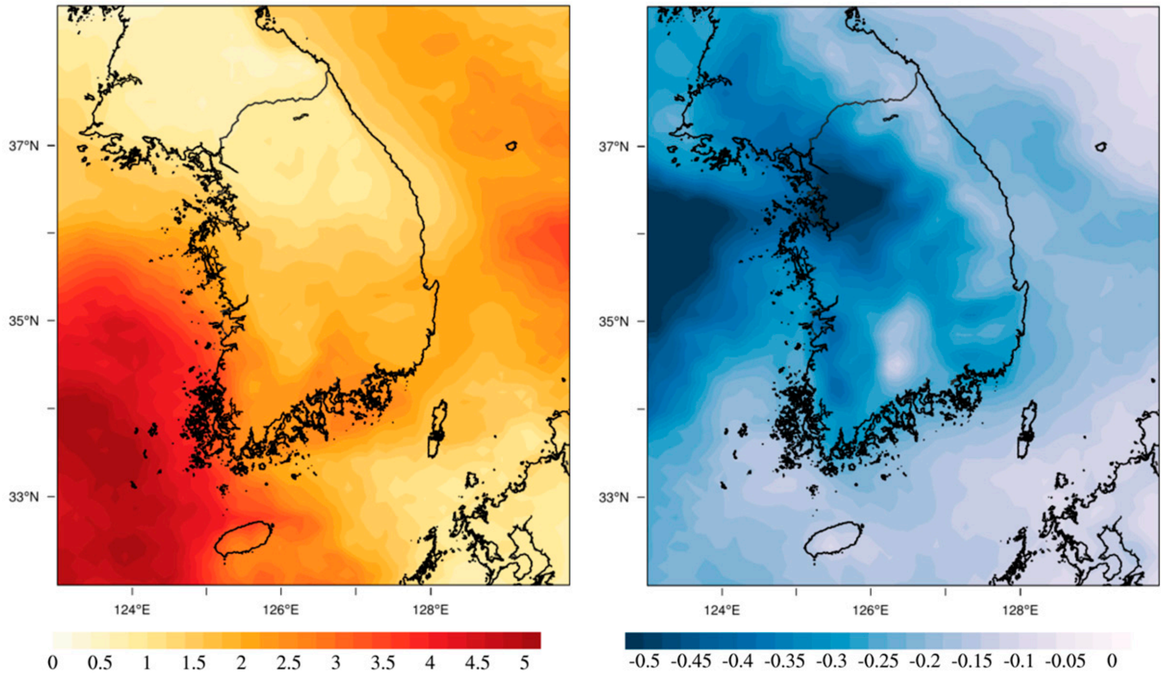


Figure 3. Changes of SO_4^{2-} and $PM_{2.5}$ concentrations (μg m^{-3}) in South Korea due to the reduction of overseas SO_2 emissions ((R_FSO2)–simulations that applied basic emissions (BASE)). (**a**) SO_4^{2-}, (**b**) $PM_{2.5}$.

The $PM_{2.5}$ concentration difference between the two experiments was clear around the Seoul Metropolitan Area (SMA) and its west coast, unlike the result of SO_4^{2-}, and showed a concentration decrease of 0.97 μg m^{-3} on average. Interestingly, despite the concentration of SO_4^{2-}, a component of $PM_{2.5}$, decreased by an average of 2.56 μg m^{-3}, the decrease in the concentration of $PM_{2.5}$ was only 0.70 μg m^{-3}. This means that the concentration of $PM_{2.5}$ components other than SO_4^{2-} may increase despite the reduced SO_2 emissions.

The results of NO_3^- and NH_4^+ were analyzed additionally for a clearer interpretation of the difference between the SO_4^{2-} and $PM_{2.5}$ results examined above. As shown in Figure 4a, the NO_3^- concentration difference between the two experiments showed a positive value overall, and the distribution pattern was similar to the result of SO_4^{2-}. This result means that the decrease in SO_4^{2-} concentration led to the production of NO_3^-. H_2SO_4, which is produced by SO_2 oxidation reactions (R1 to R3), reacts with NH_3 in the atmosphere and produces $(NH_4)_2SO_4$ (R4). Here, if the H_2SO_4 concentration decreases, the surplus NH_3 concentration increases, which in turn can promote NO_3^- production by reaction with HNO_3 (R5). Through this mechanism, the increase in NO_3^- concentration was clearly visible in the southwestern region of the domain where the decrease in SO_4^{2-} concentration was most prominent.

On the other hand, NH_4^+ concentration is mainly determined by R4 and R5, and as explained above, if SO_2 decreases, R4 decreases, and conversely, R5 increases. Therefore, the difference of NH_4^+ concentration (R_FSO2–BASE) shown in Figure 4b was negative overall, which was determined by the net effect of R4 decrease and R5 increase. These results suggest that the impact of the reduced foreign SO_2 emissions is not limited to the decrease in SO_4^{2-} concentration; it also affects concentration changes of NO_3^- and NH_4^+ through a series of chemical reactions. When the sum of differences in the SO_4^{2-}, NO_3^-, and NH_4^+ results between the two experiments was calculated (Figure 5), it was very similar to the difference of $PM_{2.5}$ results shown in Figure 3b. This means that the $PM_{2.5}$ concentration changes in South Korea due to the reduction of foreign SO_2 emissions are well explained by the concentration changes of SO_4^{2-}, NO_3^-, and NH_4^+, which are major inorganic $PM_{2.5}$ species.



(**a**) (**b**)

Figure 4. Changes of NO_3^- and NH_4^+ concentrations (μgm^{-3}) in South Korea due to the reduction of overseas SO_2 emissions (R_FSO2–BASE). (**a**) NO_3^-, (**b**) NH_4^+.

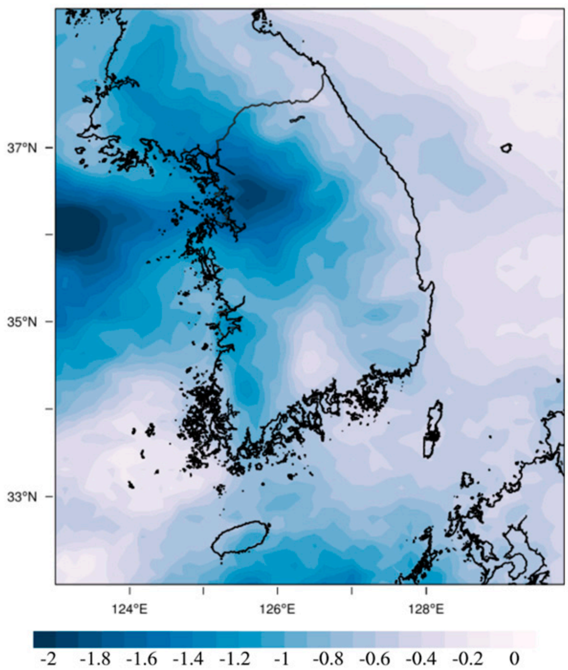


Figure 5. Changes of the sum of SO_4^{2-}, NO_3^-, and NH_4^+ concentrations (SO_4^{2-} + NO_3^- + NH_4^+, μg m^{-3}) in South Korea due to the reduction of SO_2 emissions from foreign sources (R_FSO2–BASE).

3.3. The Difference in Aerosol Direct Radiative Effect in South Korea Due to the Reduced Foreign SO_2 Emissions

The ADRE difference between the two experiments was calculated by the same method as in the previous studies [12,13] to examine the impact of SO_4^{2-} and $PM_{2.5}$ concentration differences by the foreign SO_2 reduction on the ADRE distribution changes in South Korea. To consider only the pure aerosol feedback effects, rainy and cloudy days were excluded and the ADRE difference between the two experiments was analyzed for clear-sky days only.

As shown in Figure 6, the ADRE difference between the two experiments (R_FSO2–BASE) showed negative values in the entire domain and a decrease of 0.31 Wm^{-2} on average. Particularly, the ADRE difference was large in the southwest coast of South Korea and smaller in inland regions where the domestic emissions were concentrated. Interestingly, the distribution of the ADRE difference was not consistent with the distribution of the $PM_{2.5}$ difference (Figure 3b); it was very similar to the distribution of SO_4^{2-} concentration difference (Figure 3a). This supports the research results of Yoo et al., [13], which concluded that the feedback effects of aerosol were not simply proportional to the $PM_{2.5}$ concentration but were affected by the concentration and distribution of its components, particularly SO_4^{2-}. The reason why the influence of SO_4^{2-} on the changes of the ADRE is relatively large compared to other substances is estimated to be related to the altitude where SO_4^{2-} is distributed in South Korea. The distribution of SO_4^{2-} concentration in South Korea is greatly affected by foreign sources, and the transport of SO_4^{2-} occurs mainly below the height of 2–3 km (Figure 7). Therefore, the distribution altitude of SO_4^{2-} may be relatively higher than that of other $PM_{2.5}$ components (NO_3^-, etc.), which is greatly affected by the emissions from domestic sources.

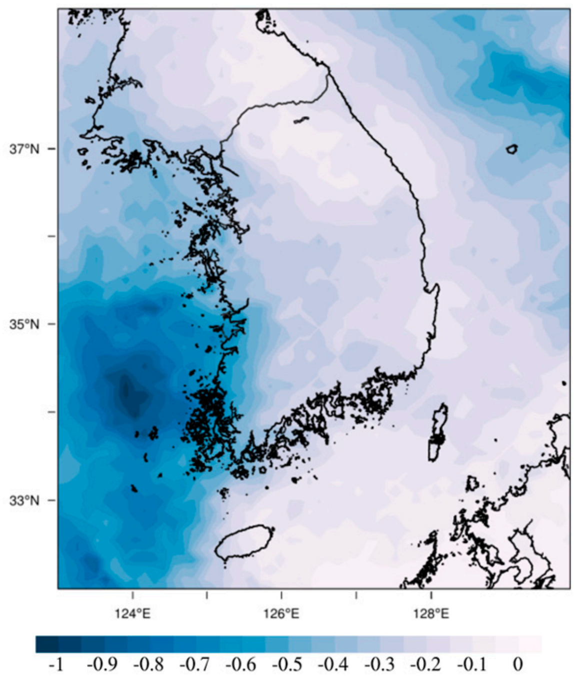


Figure 6. The changes in aerosol direct radiative effects (ADRE, Wm^{-2}) in South Korea due to the reduction of SO_2 emissions from foreign sources (R_FSO2–BASE).

Table 1 shows the concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ by altitude (the mean values of the entire domain), and the concentration of SO_4^{2-} is maintained high in the upper layer while the concentration of other substances decreases rapidly as the altitude increases. These results mean that compared to other substances, SO_4^{2-} may have a favorable condition to induce the feedback effects by reacting with short-wave solar radiation. However, it is difficult to sufficiently explain the high correlation of the ADRE and SO_4^{2-} with only the altitude difference. Therefore, an additional investigation should be conducted to determine other causes through follow-up studies.

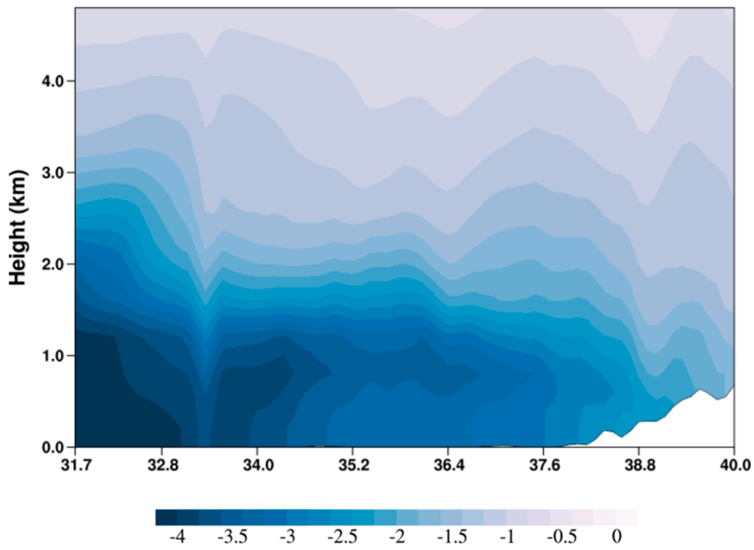


Figure 7. Vertical cross-section of SO_4^{2-} concentration difference between BASE and R_FSO2 experiments.

Table 1. The distribution of SO_4^{2-}, NO_3^-, and NH_4^+ concentrations (μg m^{-3}) according to altitude.

Altitude	SO_4^{2-}	NO_3^-	NH_4^+
Surface	6.63	6.73	4.09
100 m	6.60	6.54	4.02
500 m	6.59	5.28	3.55
1000 m	5.86	3.27	2.69
1500 m	4.44	1.40	1.63

When the above results are pieced together, it is clear that the concentration changes of $PM_{2.5}$ caused by the foreign emission reduction were determined in a complex way by the changes of SO_4^{2-}, NO_3^-, and NH_4^+, but the changes of the ADRE were determined mainly by the changes of SO_4^{2-}. Nevertheless, because the concentration change of SO_4^{2-} in South Korea depends on the inflow direction of foreign emissions into South Korea, the ADRE distribution of South Korea is not always consistent but can vary depending on the seasonal difference of long-range transport pattern. However, additional studies on various cases need to be conducted to generalize these results.

4. Conclusions

Using the WRF–CMAQ two-way coupled model, this study analyzed the impact of foreign SO_2 emission changes on the ADRE in South Korea. The differences between the BASE experiment using the basic emissions and the R_FSO2 experiment using a 50% reduction of foreign SO_2 emissions were analyzed to quantitatively examine the differences of SO_4^{2-} and $PM_{2.5}$ concentrations, and the ADRE in South Korea according to the difference in foreign SO_2 emissions.

The difference of results between the two experiments (R_FSO2–BASE) was analyzed, and the results confirmed that the inflow of SO_2, the main precursor of SO_4^{2-}, into South Korea decreased due to the reduction in the foreign SO_2 emissions. Furthermore, the results showed a clear decrease in SO_4^{2-} concentration mainly in the southwest coast of South Korea. The impact of foreign SO_2 emission reduction was not just limited to the decrease in SO_4^{2-} concentration; it also affected the concentration changes of NO_3^- (increase) and NH_4^+ (decrease) through a series of chemical reactions. The difference in $PM_{2.5}$ concentration in South Korea due to the reduction of foreign SO_2 emissions was different from the difference in SO_4^{2-} concentration, and this result shows that the change of $PM_{2.5}$ concentration is not determined simply by the change of SO_2 emissions. The changes of $PM_{2.5}$

concentration was determined in a complex way by the change of SO_4^{2-} concentration due to the change in SO_2 concentration, and the subsequent changes in NO_3^- and NH_4^+ concentrations.

The differences in SO_4^{2-} and $PM_{2.5}$ concentrations due to the reduction of foreign SO_2 emissions affected the ADRE distribution changes in South Korea. The distribution of the ADRE difference between the two experiments was not consistent with the distribution of $PM_{2.5}$ difference, but it was very similar to the distribution of SO_4^{2-} concentration difference (i.e., large in the southwest coast and the east coast of South Korea). These results support the results of Yoo et al. [13], which demonstrated that the ADRE is not simply proportional to $PM_{2.5}$ concentration but is affected by the concentration and distribution of SO_4^{2-}. The reason why SO_4^{2-} has a greater impact on the ADRE than other $PM_{2.5}$ components is that high concentration is relatively maintained in the upper layer. However, follow-up studies should be conducted for further analysis.

The results of this study confirm that the ADRE changes in South Korea may vary by region depending on the changes in foreign emissions. However, additional modeling and analysis of various cases should be conducted for further generalization since the conclusion of this study was derived through the analysis of a specific case (February 2015).

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