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Understanding the physical mechanisms of $PM_{2.5}$ formation in Seoul, Korea: assessing the role of aerosol direct effects using the WRF-CMAQ model

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Abstract

This study evaluated the impact of the aerosol direct effect (ADE) on the meteorological conditions and air quality in Seoul, South Korea (Korea), using the WRF-CMAQ coupled model. Two experiments were conducted: NF simulation without the ADE feedback and YF simulation with the ADE feedback. The ADE resulted in a decrease in shortwave radiation at the surface and an increase in shortwave radiation in the atmosphere. Additionally, the 2-m temperature, 10-m wind speed, and planetary boundary layer (PBL) height decreased. The changes in meteorological conditions due to ADE-induced atmospheric stability and restrained vertical mixing resulted in deteriorated air quality. During the simulation, $PM_{2.5}$ concentrations increased due to ADE, with daily averages rising by 4.2 $\mu g \cdot m^{-3}$ on transport day and 3.2 $\mu g \cdot m^{-3}$ on accumulation day. Process analysis (PA) was employed to investigate contributions of physical/chemical processes affecting ADE. Local emissions, aerosol processes, and horizontal advection were identified as key factors in $PM_{2.5}$ increases in Seoul. Differences in the contributions of processes were noted between transport and accumulation day due to ADE considerations. On transport day, the YF simulation exhibited a higher influx of $PM_{2.5}$ through horizontal advection, attributed to transport from upwind regions. Conversely, on accumulation day, horizontal advection decreased due to enhanced atmospheric stability, while vertical transport increased from restrained vertical mixing induced by ADE. Thus, ADE emphasized the contribution of processes to $PM_{2.5}$. Emissions and horizontal advection were primary contributors on transport day, while the emissions process dominated on accumulation day.

Keywords $PM_{2.5}$ · Transport · Accumulation · Aerosol direct effect · CMAQ

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Introduction

South Korea (hereafter, Korea) faces severe air pollution issues owing to rapid industrialization and urbanization. Among various air pollutants, $PM_{2.5}$, which refers to fine particles with a diameter of 2.5 micrometers or less, is particularly known to have detrimental effects on human health. Air quality in East Asia, where the prevailing westerlies dominate, is influenced by emissions, meteorological conditions, and long-range transport between countries (Seo et al. 2017; Liu et al. 2019; Hu et al. 2021). In particular, high $PM_{2.5}$ concentrations during the winter season in Korea are significantly influenced by long-range transport upwind regions (foreign countries) (Ji et al. 2013; Kim et al. 2017a; Choi et al. 2019; Kumar et al. 2021). Long-range transport refers to the significant influence of the transboundary transport of emissions from upwind regions along the upper layer (Lee et al. 2019a, b).

Synoptic circulation, such as high-pressure or low-pressure systems, transports air pollutants that lead to their accumulation when air stagnation occurs due to weak winds under high-pressure system conditions (Liu et al. 2013, 2021; Gao et al. 2015; Lee et al. 2019a; Yoo et al. 2022; Jun and Gu 2023). Air stagnation is characterized by reduced wind speeds resulting in poor air circulation. Recently, Korea has shown a tendency of decreasing wind speeds, indicating a correlation between severe air pollution and meteorological conditions characterized by air stagnation (Kim et al. 2017b). Due to air stagnation, air pollutants cannot disperse, leading to their accumulation in surrounding areas and further deterioration of air quality (Sun et al. 2015; Wang et al. 2018). Air stagnation currents can intensify long-range transport-induced air pollution (Jun and Gu 2023).

Aerosols in the atmosphere can scatter/absorb and reflect solar radiation, thereby influencing the Earth's radiation budget. This is referred to as the aerosol direct effect (ADE). The ADE leads to a reduction in solar radiation reaching the surface, resulting in decreased surface temperature, wind speed, and the planetary boundary layer (PBL) height (Qiu et al. 2017; Li et al. 2020). As a result of these meteorological changes, the near-surface atmosphere becomes more stable, leading to the accumulation of air pollutants near the surface and deterioration in air quality (Jung et al. 2019; Yoo et al. 2019, 2020; Huang and Ding 2021). The feedback effect between aerosols and unfavorable meteorological conditions for atmospheric dispersion can serve as a key mechanism for air pollution and further exacerbate surface air quality (Zhong et al. 2017a, b; Zhang et al. 2020). In particular, Zhong et al. (2019), through the analysis of observational data, reported that during the cumulative winter stage in Beijing, more than 70% of the increase in $PM_{2.5}$ concentrations was attributed to the influence of meteorological feedback.

However, previous studies have primarily focused on investigating the net concentration of $PM_{2.5}$, owing to the ADE, thus lacking a comprehensive assessment of the contributions of physical/chemical processes to concentration changes. In particular, it is necessary to quantitatively analyze the contributions of physical/chemical processes, such as horizontal and vertical transport and local sources, to $PM_{2.5}$ formation mechanisms in the Korean Peninsula, with its complex topography and associated diverse scales of meteorological phenomena. Gao et al. (2015) using process analysis revealed that during heavy haze episodes in winter, advection process contributed up to 70% to $PM_{2.5}$ concentrations in Hangzhou. The $PM_{2.5}$ formation mechanisms, including long-range transport, accumulation, and dispersion of air pollutants, may depending on synoptic circulation pattern. Therefore, it is necessary to investigate the ADE with respect the $PM_{2.5}$ formation mechanisms on the Korean Peninsula.

This study aimed to evaluate the influence of ADE on the $PM_{2.5}$ formation mechanism in Seoul, Korea. We utilized the WRF-CMAQ two-way coupled model to account for the influence of ADE and conducted a quantitative analysis of the contributions of physical and chemical processes, focusing on the formation mechanism of $PM_{2.5}$.

Materials and methods

Modeling system

This study used the WRF-CMAQ two-way coupled model (WRF version 3.8 and CMAQ version 5.2), which was developed to consider the ADE through the aerosol feedback option (Wong et al. 2012). Figure 1 shows the modeling domain covering South Korea and Northeast Asia, with a horizontal resolution of 12 km (250 × 230) and 40 vertical layers. The initial and boundary conditions for the WRF model were derived from the Final Operational Global Analysis reanalysis data with a horizontal resolution of 0.25° × 0.25° provided by the National Centers for Environmental Prediction. The physics options consisted of the WRF single-moment 5-class scheme microphysics scheme (WSM5; Hong et al. 2004), Pleim-Xiu land surface model (Xiu and Pleim 2001), RRTMG shortwave and longwave scheme (Iacono et al. 2008), Asymmetric Convective Model version 2 (ACM2; Pleim 2007a; Pleim 2007b) PBL scheme, and Kain–Fritsch cumulus parameterization (Kain 2004).

CMAQ modeling domain covering South Korea and Northeast Asia, with a horizontal resolution of 12 km (244 × 224). The initial and lateral boundary conditions for CMAQ modeling used to default profile. We used the Mosaic Asian Anthropogenic Emission (MIX) (Li et al. 2017) with a horizontal resolution of 0.25° × 0.25° for the CMAQ model in Asia and the Clean Air Policy Support System (CAPSS) emission provided by the National Institute of Environmental Research (NIER) in South Korea. Additionally, we used the Model of Emissions of Gases and Aerosols from Nature (MEGAN version 2.1) for vegetation emissions. We used the Carbon Bond 05 mechanism with updated toluene chemistry (CB05tucl) for the gas phase (Whitten et al. 2010) and AERO6 for the aerosol chemical mechanisms (Nolte et al. 2015). We conducted two experiments: a base simulation without aerosol feedback (NF) and one with aerosol feedback (YF). The difference between the NF and YF simulation results was used to analyze the impact of the ADE.

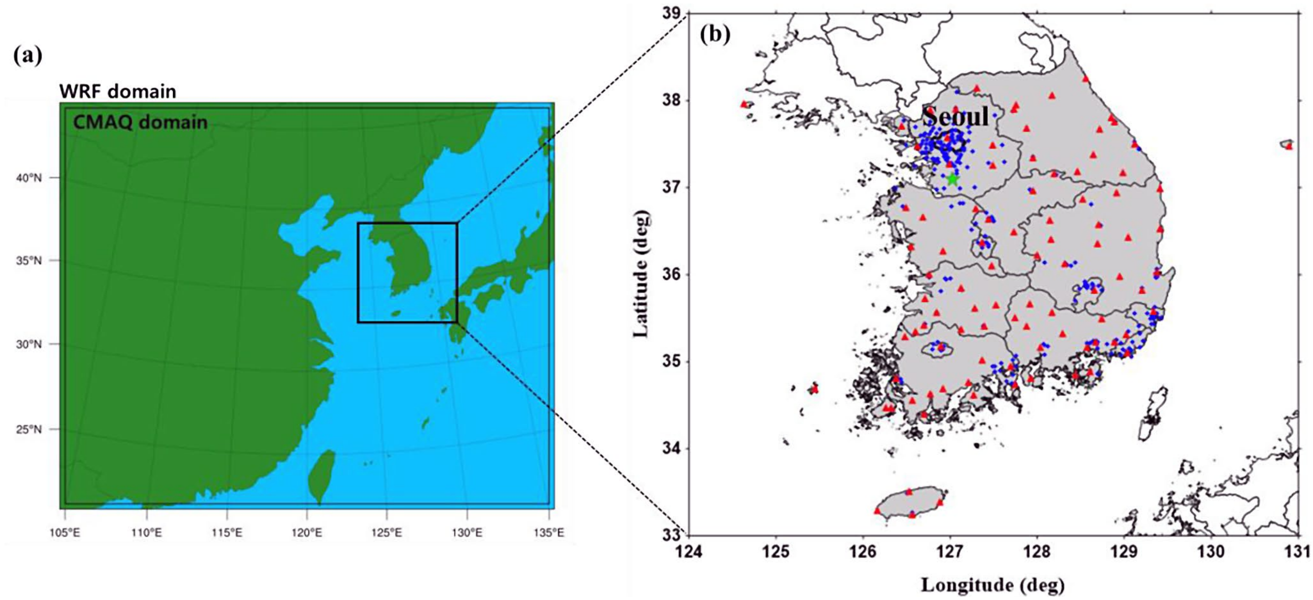

Fig. 1 **a** Study modeling domain for simulations and **b** locations of meteorological observation (red triangles), Osan sounding observation (green star), and PM2.5 concentrations measurements (blue dots) stations

Integrated process rate (IPR) analysis

The Integrated Process Rate (IPR) of the process analysis (PA) module of the CMAQ model was used to investigate the contribution of each physical and chemical process to $PM_{2.5}$ (Gipson and Young 1999). IPR analysis can quantify the net effects of physical and chemical processes on the formation and reduction of gas and aerosol species. An analysis of these processes can reveal the major sources or sinks of $PM_{2.5}$. IPR analysis calculates the contributions of individual processes: horizontal advection (HADV), horizontal diffusion (HDIF), vertical advection (ZADV), vertical diffusion (VDIF), emission (EMIS), aerosol processes (AERO), cloud processes (CLDS), and dry deposition (DDEP). In this study, we analyzed vertical transport (ZTRA = ZADV + VDIF) by considering the combined inflow and outflow of vertical advection and diffusion. In addition, horizontal diffusion (HDIF) was not analyzed in this study because its impact on $PM_{2.5}$ concentrations was minimal compared with that of horizontal advection (Seaman et al. 2000).

Simulation episode

To understand the impact of ADE on $PM_{2.5}$, we selected and simulated a pollution episode that occurred during the winter of 2017. On 18 and January 19, 2017, in Seoul, the daily average $PM_{2.5}$ concentrations exceeded 70 $\mu g \cdot m^{-3}$, and the hourly average exceeded 100 $\mu g \cdot m^{-3}$. During the simulation period, an analysis of the synoptic weather conditions revealed that on January 17, a low-pressure system was located to the west of the Korean Peninsula (Fig. 2). This low-pressure system subsequently moved northeastward, and, the Korean Peninsula was dominated by the high-pressure system on January 18. The influence of high-pressure systems accompanied by westerly or north-westerly winds can result in the transport of air pollutants, including $PM_{2.5}$, from upwind regions to downwind regions. Consequently, this process can exacerbate the air quality over the Korean Peninsula. On January 18, the polluted air was transported to the Korean Peninsula, increasing the $PM_{2.5}$ concentrations, with the daily average exceeding 72 $\mu g \cdot m^{-3}$. On January 19, as the high-pressure system expanded, the influence of weak winds (1.3 $m \cdot s^{-1}$ in Seoul meteorological station) and air stagnation due to the high-pressure system may lead to the accumulation of transported air pollutants ($PM_{2.5}$). The daily average $PM_{2.5}$ concentrations were over 73 $\mu g \cdot m^{-3}$ in Seoul. Therefore, Seoul on the Korean Peninsula is considered suitable for analyzing the impact of the ADE on the formation mechanisms of $PM_{2.5}$ during this episode period. This is due to the high concentrations of $PM_{2.5}$ that can occur as a result of long-range transport from upwind regions and the accumulation of pollutants caused by air stagnation. The modeling period was set from January 15 to 22, 2017, including the spin-up time, and the analysis was performed by focusing on January 18 and 19.

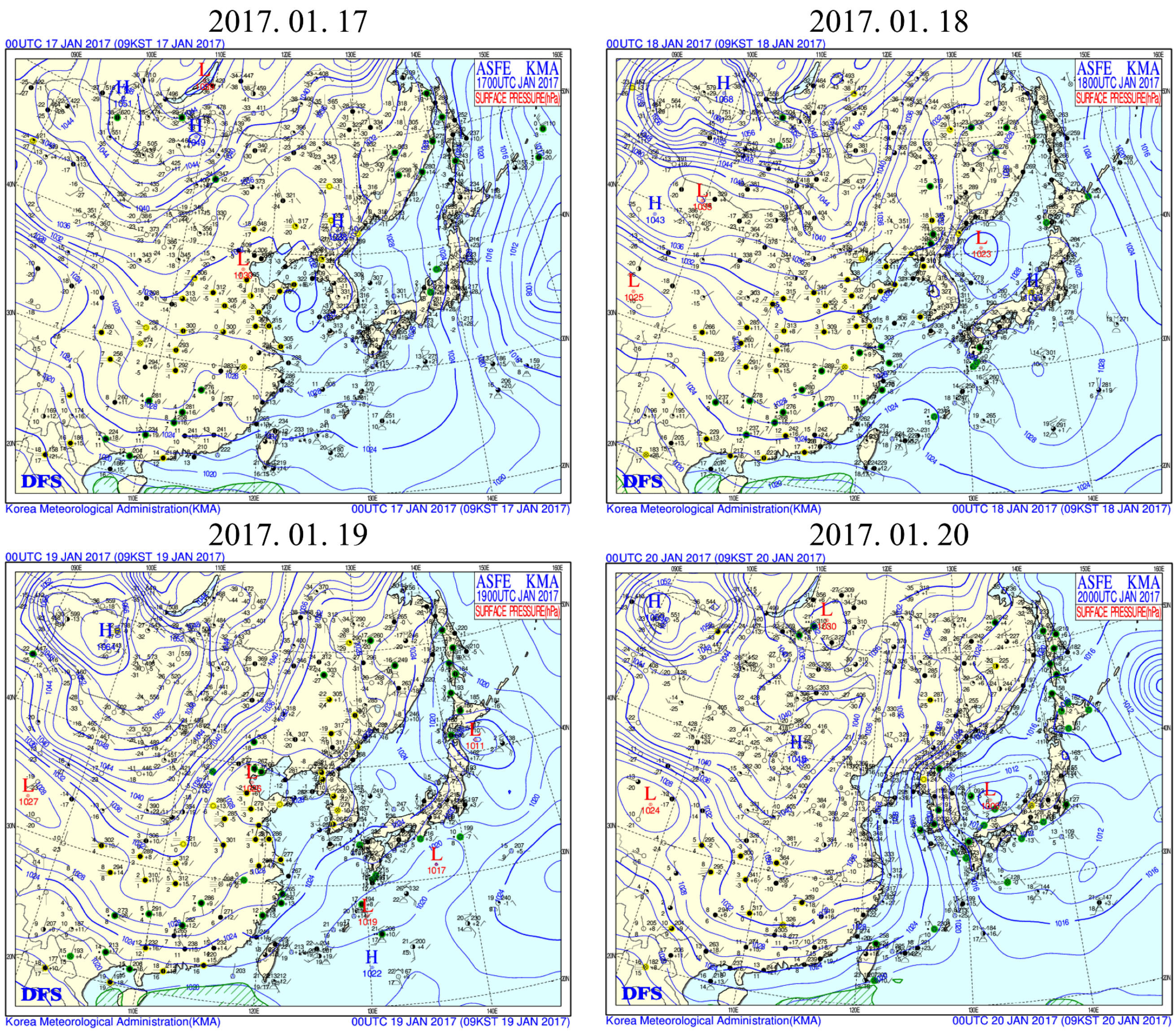

Fig. 2 Surface synoptic weather map for January 17–20, 2017, at 0900 LST

Results

Model evaluation

To evaluate the performance of the WRF model, we compared the observed and simulated NF simulation values of 2-m temperature (T2) and 10-m wind speed (WS10) at 95 Automated Surface Observing System (ASOS) meteorological observation stations (red triangles in Fig. 1) in South Korea (Fig. S1).

For temperature (T2), the WRF model captured its diurnal and daily variations with index of agreement (IOA) and correlation coefficient (R) values of 0.93 and 0.87, respectively, but marginally overestimated these with a mean bias (MB) of 0.40 °C. For wind speed (WS10), the results were marginally overestimated (MB 1.43 $m \cdot s^{-1}$) compared with the observed values. The IOA and R values were 0.72 and 0.65, respectively, indicating good agreement. Although discrepancies exist, the model performed well overall and satisfied the baseline suggested by Emery et al. (2001) (T2: Bias $\leq \pm 0.5$ °C; IOA ≥ 0.7, WS10: Bias ≤ 2.0 $m \cdot s^{-1}$; IOA ≥ 0.6). Furthermore, the temperature profiles in the NF simulation closely resembled sounding observations, and effectively simulated the inversion layer below 900 hPa on January 19 (Fig. S4). On January 18, the wind speed profile was overestimated by 2 $m \cdot s^{-1}$ near the surface and underestimated between 700 and 900 hPa. The YF simulation, considering the ADE, showed a minor improvement in accuracy; detailed information is presented in Table S1.

To evaluate the results of the CMAQ model (YF simulation), we measured $PM_{2.5}$ concentrations at 256 Air Quality Monitoring Stations (AQMS) in South Korea (blue circles in Fig. 1). Figure 3 shows the time series of the observed and simulated $PM_{2.5}$ concentrations. Both the measured and simulated $PM_{2.5}$ concentrations showed an increasing trend from January 17 and decreased from January 20. Although the simulation results tended to underestimate the measured values marginally, the CMAQ model captured the temporal patterns of $PM_{2.5}$ concentrations with a high IOA (0.76). While in Seoul, the CMAQ model exhibited a slight tendency to underestimate the measured $PM_{2.5}$ concentrations, the performance of the CMAQ model results appeared similar for $PM_{2.5}$ concentrations in the Korea and Seoul (Fig. S2). The inaccuracies in the CMAQ model results can be attributed to uncertainties in the emission data, initial/boundary conditions, and meteorological model results (Miao et al. 2017). However, the model evaluation results from the CMAQ model also indicated a minor improvement in the performance of the YF simulation compared with the NF simulation.

Horizontal distribution of $PM_{2.5}$ concentrations

The horizontal distribution of $PM_{2.5}$ concentrations from the NF simulation is shown in Fig. 4. On January 17 (2100 LST), $PM_{2.5}$ showed a high concentration (200 $\mu g \cdot m^{-3}$) in inland China and a relatively low concentration in the Korean Peninsula. On January 18, influenced by the northwesterly winds, the high $PM_{2.5}$, which was transported to the Korean Peninsula, increased compared with the previous day. Subsequently, weak winds (average daily wind speed in Seoul 1.6 $m \cdot s^{-1}$) appeared on the Korean Peninsula. Weak winds can increase $PM_{2.5}$, owing to the weaker advection and stable atmospheric conditions. Due to the accumulation of transported $PM_{2.5}$ from the previous day and local anthropogenic emissions (Fig. S2), high-level concentrations exceeding 80 $\mu g \cdot m^{-3}$ persisted in the western part of the Korean Peninsula on January 19. Air quality improved overall due to the spread of $PM_{2.5}$ to the southeast by strong northwesterly winds. The horizontal distribution results showed high concentrations on January 18 and 19: however, the primary mechanisms for $PM_{2.5}$ formation varied. On January 18, transport by northwesterly winds appeared to contribute to high concentrations of $PM_{2.5}$. However, on January 19, due to inversion layer in the lower atmosphere (Fig. S4), pollutants emitted from locally from anthropogenic sources were unable to disperse into the surrounding region and resulted in high concentrations due to accumulation. Therefore, based on the formation mechanism of $PM_{2.5}$, we divided the simulation period into a transport day on January 18 and an accumulation day on January 19.

Aerosol direct effect (ADE) on $PM_{2.5}$ formation mechanisms

Aerosols affect the radiation at the surface and in the atmosphere by absorbing or reflecting incoming solar radiation, or both. Figure 5 shows the impact of ADE on shortwave radiation at the surface and in the atmosphere, calculated as the difference between the results of the YF and NF simulation results. As expected, the ADE resulted in a reduction in surface shortwave radiation and an increase in atmospheric radiation. In particular, the influence of the ADE was more pronounced in the southwest–northeast regions on transport days and in the western part of the Korean Peninsula on accumulation days with higher aerosol ($PM_{2.5}$) loading. The shortwave radiation at the surface (SW_SFC) averaged over Seoul was reduced by 13.2 $W \cdot m^{-2}$ (13.6%) during the transport day and 21.5 $W \cdot m^{-2}$ (17.2%) during the accumulation day. On the other hand, the shortwave radiation in the atmosphere (SW_ATMOS) increased by an average of 11.5 $W \cdot m^{-2}$ (33.2%) during the transport day and 9.9 $W \cdot m^{-2}$ (32.9%) during the accumulation day (Table 1).

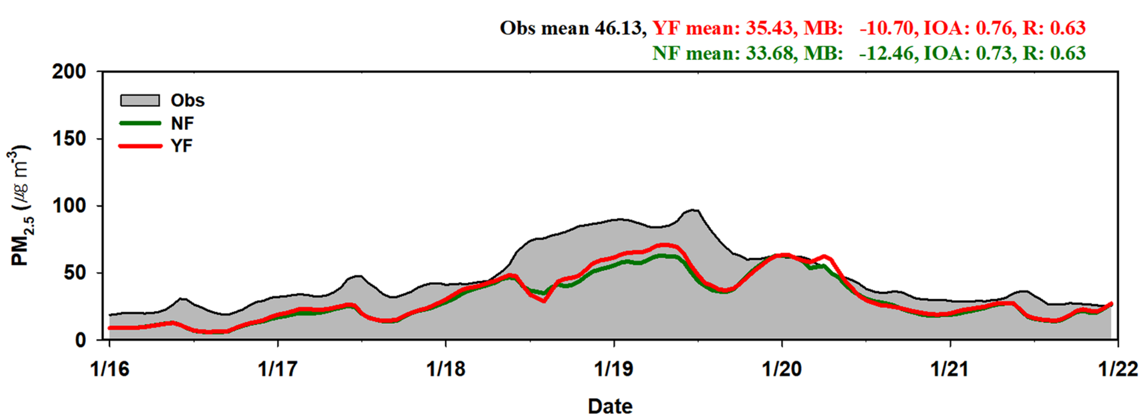

Fig. 3 Time series of measured and simulated (YF and NF simulations) $PM_{2.5}$ concentrations. MB, IOA, and R represent mean bias, index of agreement, and correlation coefficient, respectively

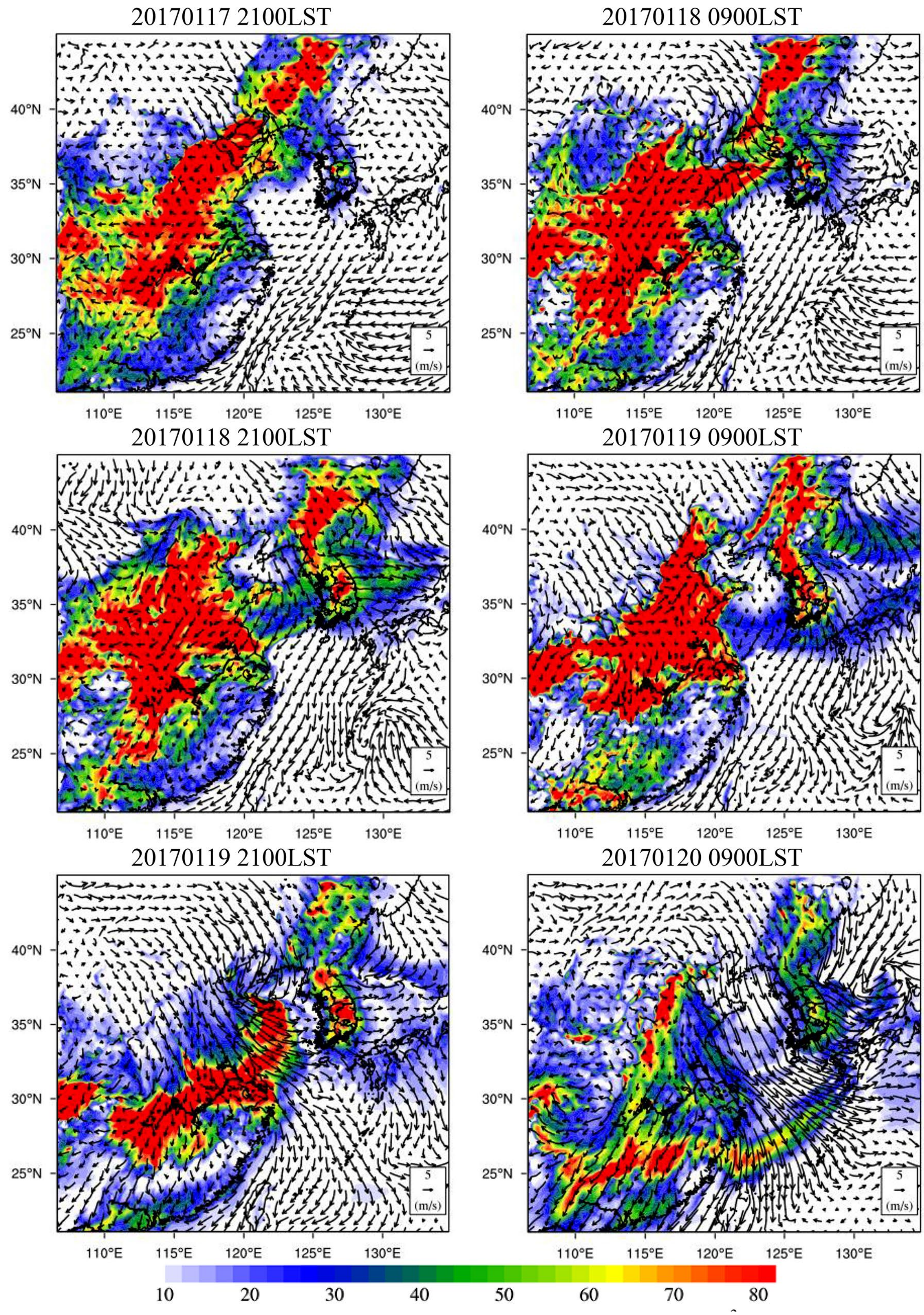

Fig. 4 Horizontal distribution of surface $PM_{2.5}$ concentrations ($\mu g \cdot m^{-3}$) and wind vectors (arrows, $m \cdot s^{-1}$) for NF simulation

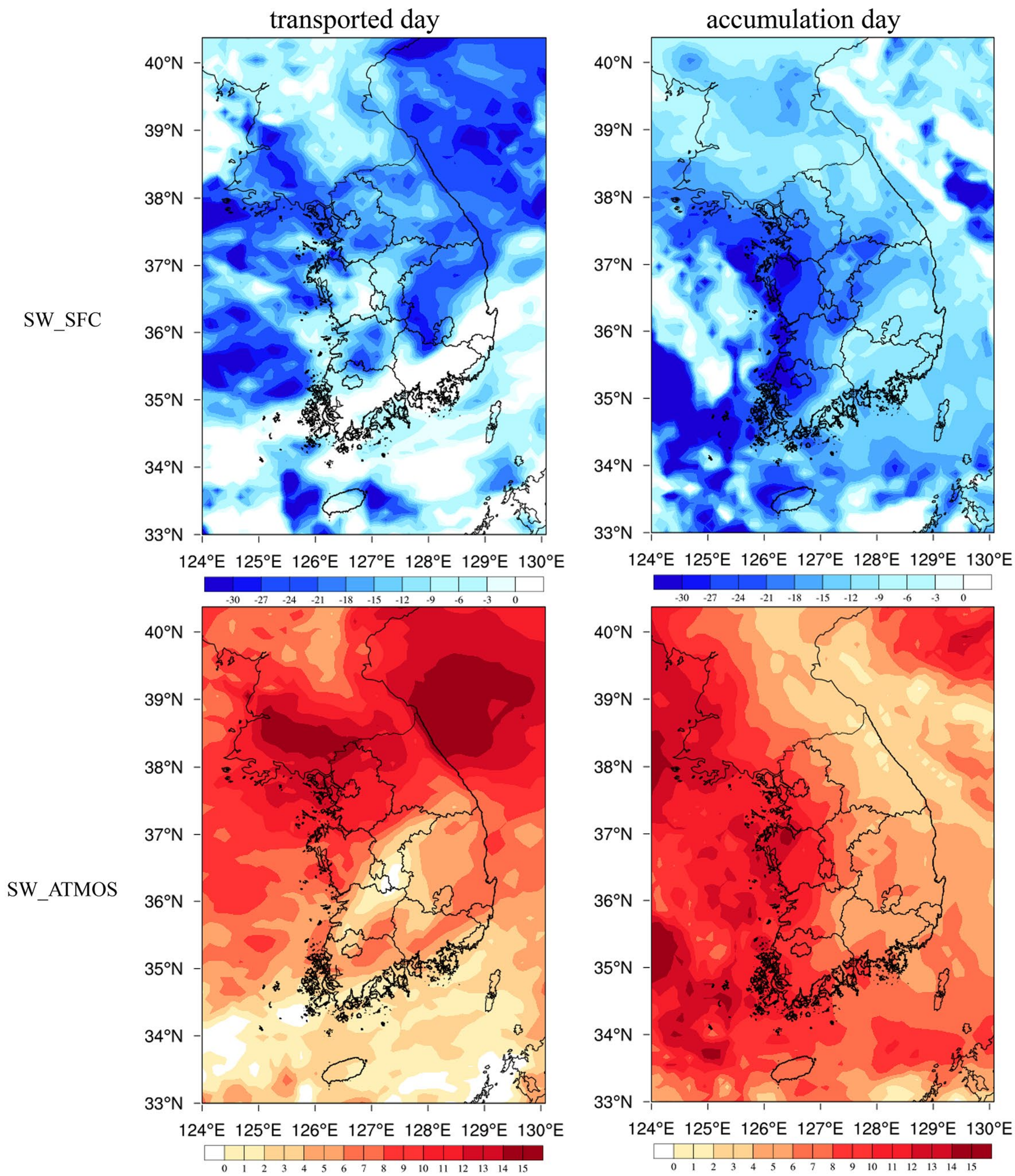

Fig. 5 Horizontal distribution of the aerosol direct effects (YF minus NF simulation) on shortwave radiation (W $m_{-2.5}$)at the surface (SW_SFC) and in the atmosphere (SW_ATMOS)

The ADE affects shortwave radiation, in turn affecting meteorological conditions, such as air temperature, wind speed, and PBL height. Reductions in surface shortwave radiation can inhibit surface heating, decreasing air temperature and PBL height. In Seoul, the average 2-m temperature showed no significant change on the transport day but a decrease of 1.5 ℃ (from −3.3 to −4.8 ℃) on accumulation days (Table 1). The average 10-m wind speed showed a similar decrease of 0.2 $m \cdot s^{-1}$ on both days. Surface cooling by ADE can also inhibit PBL development (Zhong et al. 2019; Su et al. 2020). The daytime PBL heights decreased to 120-m (from 576 to 456-m) during the transport day and to 92-m (from 630 to 538-m) during the accumulation day (Table 1).

The decrease in temperature, wind speed, and PBL height due to the ADE stabilized the atmosphere, creating favorable meteorological conditions for the accumulation of air pollutants. Atmospheric stabilization by ADE weakens the advection, diffusion, and vertical mixing of air pollutants, which can lead to increased concentrations due to their accumulation.

Table 1 Daily average $PM_{2.5}$ concentrations from NF simulation and aerosol direct effects on meteorological conditions for shortwave radiation at the surface (SW_SFC) and in the atmosphere (SW_ATMOS), 2-m temperature (T2), 10-m wind speed, planetary boundary layer (PBL) height, $PM_{2.5}$ concentrations. The values within parentheses represent the percentage change due to the ADE compared to the NF simulation

	Transported day	Accumulation day
Daily average $PM_{2.5}$	48.0 $\mu g{\cdot}m^{-3}$	63.9 $\mu g{\cdot}m^{-3}$
SW_SFC	-13.2 $W{\cdot}m^{-2}$ (-13.6%)	-21.5 $W{\cdot}m^{-2}$ (-17.2%)
SW_ATMOS	11.5 $W{\cdot}m^{-2}$ (33.2%)	9.9 $W{\cdot}m^{-2}$ (32.9%)
T2	0.0 °C	-1.5 °C
WS10	-0.2 $m{\cdot}s^{-1}$	-0.2 $m{\cdot}s^{-1}$
PBL height	-120 m	-92 m
$PM_{2.5}$	4.2 $\mu g{\cdot}m^{-3}$ (8.8%)	3.2 $\mu g{\cdot}m^{-3}$ (4.9%)

Changes induced by the ADE were more pronounced in regions of high concentrations, with increases exceeding 50 $\mu g{\cdot}m^{-3}$ on the transport day (red box in Fig. 6). Increased $PM_{2.5}$, due to the ADE over high concentrations of upwind regions, was subsequently transported into the Korean Peninsula, resulting in a concentration increase of 4.2 $\mu g{\cdot}m^{-3}$ in Seoul. In contrast, $PM_{2.5}$ concentrations predominantly increased on the accumulation day in South Korea, with a rise of 3.2 $\mu g{\cdot}m^{-3}$ recorded in Seoul. These changes in $PM_{2.5}$ concentrations over South Korea could be attributed to a decrease in wind speed and increased atmospheric stability induced by the ADE.

In summary, when long-range transport contributed to the high concentrations in the Korean Peninsula, the ADE impact was dominated by the transport effects of increased concentrations from foreign countries. In addition, when high concentrations occurred due to accumulation, the

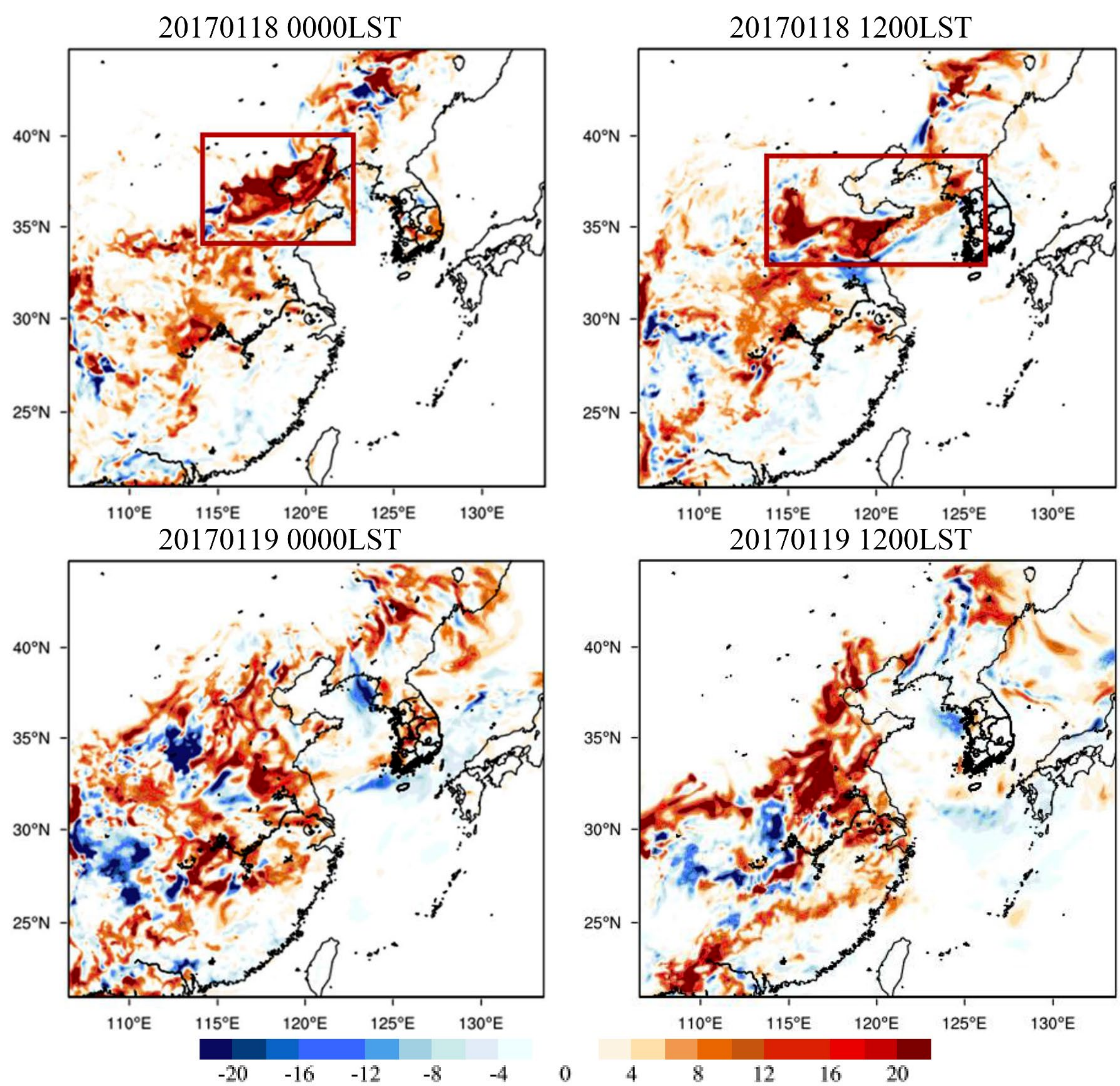

Fig. 6 Horizontal distribution of the aerosol direct effects (YF minus NF simulations) on $PM_{2.5}$ concentrations ($\mu g{\cdot}m^{-3}$) at the surface. The red box indicates areas where $PM_{2.5}$ increased by 50 $\mu g{\cdot}$ m^{-3} or more in the upwind region due to the aerosol direct effects

increase in atmospheric stability induced by the ADE led to an increase in $PM_{2.5}$ concentrations in South Korea.

Physical/chemical process contributions to $PM_{2.5}$ formation mechanisms

To quantitatively assess the formation mechanisms of $PM_{2.5}$ and the influence of ADE based on these mechanisms, we present each physical and chemical process in Seoul using IPR (Fig. 7). A positive value indicates that the inflow of $PM_{2.5}$ from this process was higher than the outflow and contributed to the increase in $PM_{2.5}$. In this study, the HDIF, CHEM, and CLDS processes were not analyzed because of their negligible impact on $PM_{2.5}$ compared with the others. The contribution of each process was determined by averaging the hourly values during the transport day or accumulation day.

The results of the NF simulation showed that the EMIS, AERO, and HADV processes were positive during the transport and accumulation days, whereas ZTRA and DDEP were negative. The results of the NF simulation showed that the emissions of primary pollutants from local sources, aerosol processes, and horizontal advection processes primarily contributed to the increase in $PM_{2.5}$. Furthermore, the ZTRA and DDEP processes contributed to the removal of $PM_{2.5}$.

Despite changes in processes due to the ADE, emissions of primary pollutants (EMIS) could be attributed to increases in $PM_{2.5}$ and dry deposition (DDEP) to decreased concentrations. However, the YF simulation revealed changes in the contribution of the ADE. In comparison to the NF simulation, the YF simulation showed a greater influx of $PM_{2.5}$ concentrations through horizontal advection (HADV + 2.30 $\mu g \cdot m^{-3}$) during the transport day. These results indicate that horizontal advection contributed to the increase in $PM_{2.5}$, which can be attributed to transport from upwind regions during the transport day. Despite the restrained vertical mixing by the ADE, decreases in ZTRA (–1.39 $\mu g \cdot m^{-3}$) were noted. The decrease in the ZTRA is explained by the vertical distribution analysis described in next Section. The enhancement of $PM_{2.5}$ concentrations (+4.2 $\mu g \cdot m^{-3}$) induced by the ADE could be mainly attributed to the contribution of the HADV process during the transport day. During accumulation days, the reduced wind speed due to the ADE resulted in a decrease in the inflow of HADV (from +2.67 to +0.23 $\mu g \cdot m^{-3}$). In addition, the restrained vertical mixing near the surface (ZTRA increased by +0.96 $\mu g \cdot m^{-3}$) and increase in secondary particle production (AERO + 0.60 $\mu g \cdot m^{-3}$) due to increased aerosol precursors and water vapor in the PBL (Chen et al. 2019) could be a major factor contributing to the increase in $PM_{2.5}$ concentrations. There was little variation in the EMIS process between the NF and YF simulations because the local emissions remained relatively constant. Additionally, a minor increase in DDEP was observed in the YF simulation. The increase in DDEP (+0.19 $\mu g \cdot m^{-3}$ during the transport day and +0.51 $\mu g \cdot m^{-3}$ during the accumulation day) was attributed to the reduction in wind speed caused by the ADE, leading to a decrease in the dry deposition. During the accumulation day, the enhancement of $PM_{2.5}$ concentrations (+3.2 $\mu g \cdot m^{-3}$) induced by the ADE could be attributed to a decrease in the HADV and an increase in the ZTRA, DDEP, and AERO processes.

The contribution analysis using IPR revealed that on the transport day, emissions (EMIS) and horizontal advection (HADV) processes contributed to the increase in $PM_{2.5}$ concentrations, while on the accumulation day, the emission (EMIS) process played a predominant role. Additionally,

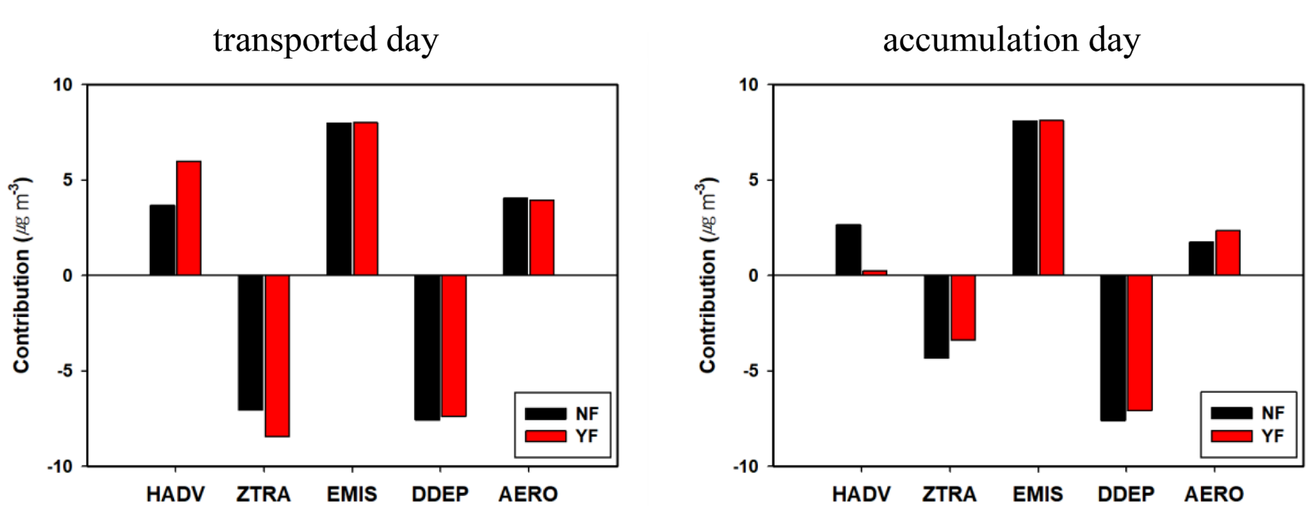

Fig. 7 Average contribution of individual processes to the $PM_{2.5}$ concentrations. HADV – horizontal advection, ZTRA – vertical transport (advection + diffusion), EMIS – emission, DDEP – dry deposition, AERO – aerosol processes

the influence of ADE highlighted the processes associated with the increase in $PM_{2.5}$ on both the transport and accumulation day.

Vertical distribution induced by aerosol direct effects

In this section, we examine the vertical distribution of meteorological conditions and $PM_{2.5}$ concentrations due to the ADE. Figure 8 shows the time series of (a) $PM_{2.5}$ for the NF simulation and PBL heights (solid black line: NF simulation, purple dashed line: YF simulation), and the difference by ADE ((b) $PM_{2.5}$ concentration, (c) u-component of wind, (d) HADV, and (e) ZTRA) over Seoul.

The results of NF simulation showed elevated concentrations above 50 $\mu g \cdot m^{-3}$ during morning and nighttime near the surface, while $PM_{2.5}$ concentrations diluted during the daytime due to enhanced vertical mixing induced by surface heating. However, the vertical distribution of $PM_{2.5}$ in the upper layer differed on the transport and accumulation days. On the accumulation day, high concentrations were observed near the surface. In contrast, on the transport day, high concentrations of 60 $\mu g \cdot m^{-3}$ occurred near 500–2000-m altitudes from dawn to noon (red box in Fig. 8a). Previous studies have demonstrated that in the case of long-range transport from foreign countries to South Korea, air pollutants are transported above the nocturnal boundary layer (Lee et al. 2019a). Therefore, high concentrations of $PM_{2.5}$ above the nocturnal boundary layer on transport days could be attributed to the long-range transport of air pollutants. In addition, the growth of the daytime boundary layer promotes the vertical mixing of transported pollutants, thereby leading to higher concentrations near the surface (Lee et al. 2019a, b). This study also showed elevated levels of $PM_{2.5}$ at altitudes ranging from 500 to 2000-m during the nighttime on the transport day, and it is believed that these high concentrations in the upper layer were transported and influenced the surface through vertical mixing (Lee et al. 2019a). In contrast, on the day of accumulation, the absence of a high-concentration core above the nocturnal boundary layer indicated that the high concentrations near the surface were caused by accumulation.

Generally, the ADE increases the concentration of air pollutants with decreasing air temperature, wind speed, and PBL) height. This study also revealed that the daily average $PM_{2.5}$ concentrations in Seoul increased by 3 to 4 $\mu g \cdot m^{-3}$ due to ADE (Table 1). However, differences were found between the transport and accumulated days in the time series of the vertical distribution. Despite the meteorological conditions that could have led to an increase in $PM_{2.5}$, such as decreased westerly winds (u-component) and restrained vertical mixing caused by atmospheric stabilization (blue box in Fig. 8b), the $PM_{2.5}$ concentrations decreased in the morning of the transport day. Westerly winds can transport air pollution from upwind regions to the Korean Peninsula. Therefore, the decrease in HADV (blue box in Fig. 8d) because of the decrease in u-component (blue box in Fig. 8c) also decreased the amount of $PM_{2.5}$ transported from foreign countries to the Korean Peninsula.

After 15:00 on the transport day, it was observed that $PM_{2.5}$ concentrations near the surface increased by more than 30 $\mu g \cdot m^{-3}$ (red box in Fig. 8b). This is attributed to the transport of $PM_{2.5}$ concentrations increased by the aerosol direct effect (ADE) in the upwind region to the Korean Peninsula in the afternoon. Additionally, the increase in $PM_{2.5}$, which was facilitated by favorable meteorological conditions for the accumulation of $PM_{2.5}$, included a decrease in the westerly winds (u-component) near the surface (blue box in Fig. 8c). Despite the restrained vertical mixing caused by the ADE, ZTRA decreased (blue box in Fig. 8e) in the afternoon on the transport day. According to Li et al. (2021), it has been reported that the aerosol feedback and the vertical exchange of $PM_{2.5}$ can enhance secondary circulation in the boundary layer. Therefore, it can be inferred that the ZTRA decreased (increased outflow-induced vertical mixing) during the transport day owing to increased upwind motion by secondary circulation. Overall, $PM_{2.5}$ concentrations increased due to the larger magnitude of horizontal transport from upwind regions compared with the decrease caused by vertical transport, in Seoul.

During the accumulation day, the impacts of the ADE resulting in different processes differed. The horizontal advection of $PM_{2.5}$ decreased (blue box in Fig. 8d) owing to the reduction of the u-component by ADE (blue box in Fig. 8c). In addition, the $PM_{2.5}$ concentrations increased near the surface (red box in Fig. 8d) owing to a decrease in the outflow of $PM_{2.5}$ by the restrained vertical mixing. Therefore, the increase in $PM_{2.5}$ concentrations during the accumulation day is attributed to weak removal (reduced outflow) by horizontal and vertical transport and secondary particle production due to aerosol chemical processes in Seoul (Fig. 7).

In this study, we examined the effect of ADE on the formation mechanism of $PM_{2.5}$ and air quality in Seoul. Considering the ADEs, the reduction in shortwave radiation, air temperature, wind speed, and PBL heights, along with increased atmospheric stability and restrained vertical mixing, led to elevated near-surface $PM_{2.5}$. However, we identified that the main processes contributing to the concentration increase varied depending on the formation mechanism of $PM_{2.5}$. During the transport day, ADE caused an accumulation of $PM_{2.5}$ concentrations near the upwind region, significantly contributing to its transportation to the Korean Peninsula and leading to a substantial increase in $PM_{2.5}$. In

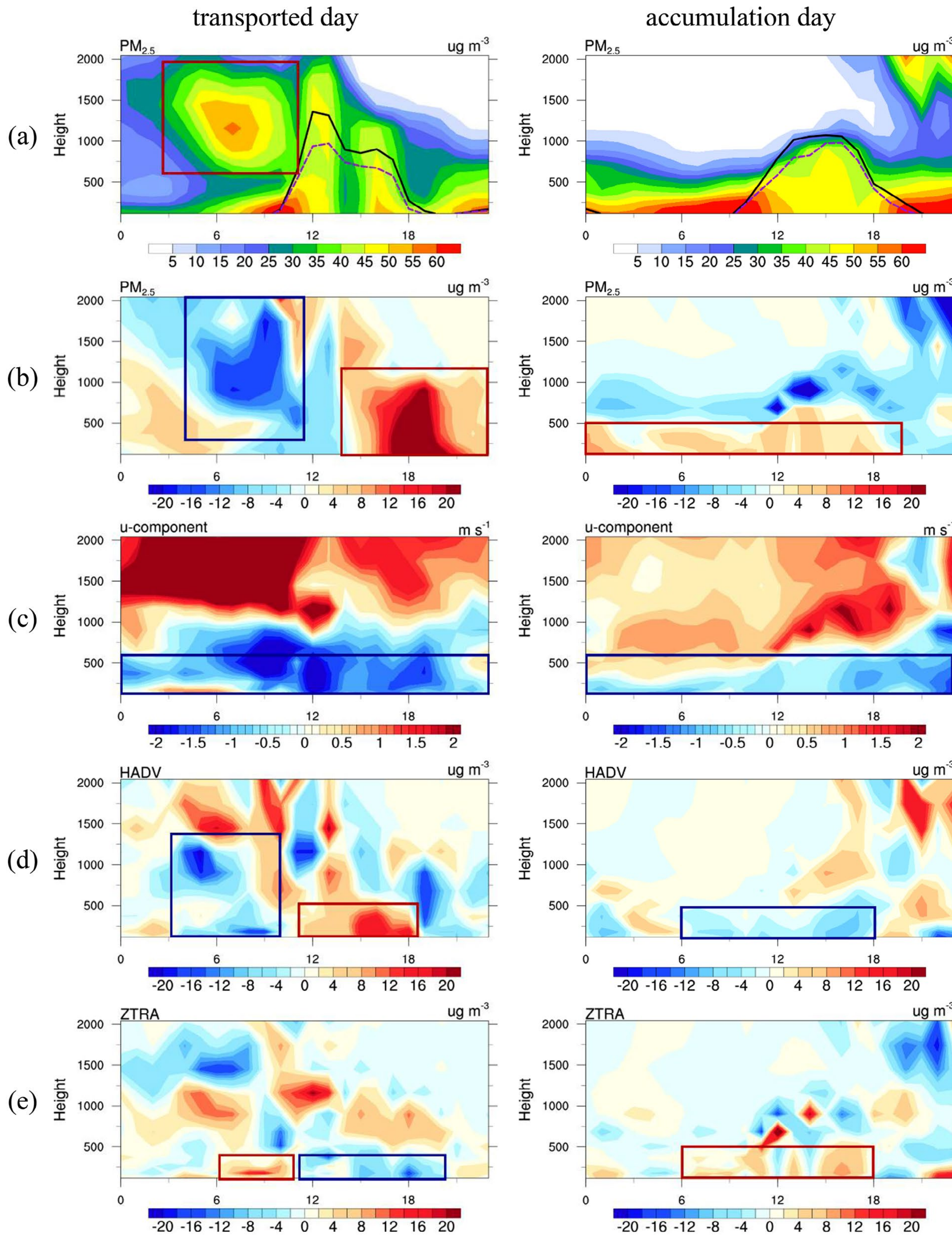

Fig. 8 Time series of time-height sections of **a** $PM_{2.5}$ concentrations ($\mu g{\cdot}m^{-3}$) from NF simulation, the difference in **b** $PM_{2.5}$ concentrations ($\mu g{\cdot}m^{-3}$), **c** u-component of wind (U, $m{\cdot}s^{-1}$), **d** horizontal advection (HADV, $\mu g{\cdot}m^{-3}$), and **e** vertical transport (ZTRA, $\mu g{\cdot}m^{-3}$) in Seoul. The red (blue) boxed areas in the figure indicate prominent increase (decrease) due to the aerosol direct effects. The solid black and purple dashed line represent the PBL heights for NF and YF simulations, respectively

contrast, during the accumulation day, the enhanced atmospheric stability induced by ADE (reduced westerly winds and PBL heights) led to a decrease in horizontal transport and restrained vertical mixing, thereby increasing $PM_{2.5}$.

Summary and conclusion

In this study, we investigated the impact of the ADE on the formation mechanism of $PM_{2.5}$ in Seoul, South Korea using the WRF-CMAQ two-way coupled model. To quantitatively analyze the $PM_{2.5}$ formation mechanism (transport and accumulation days), the contribution of physical/chemical processes was investigated using the Integrated Process Rate (IPR) of the CMAQ model's PA module. Two simulations were conducted to investigate the impact of ADE on the meteorological conditions and air quality.

Considering the ADE, the shortwave radiation at the surface decreased, and the shortwave radiation in the atmosphere increased. Furthermore, reductions in temperature, wind speed, and PBL height resulted in atmospheric stability and favorable meteorological conditions for aerosol accumulation. These changes in the meteorological conditions exacerbated air pollution during the simulation period. When considering the ADE, the daily average $PM_{2.5}$ concentrations in Seoul showed an increase of 4.2 $\mu g \cdot m^{-3}$ on the transport day and 3.2 $\mu g \cdot m^{-3}$ on the accumulation day, particularly in regions with high $PM_{2.5}$ concentrations.

During the simulation period, it was revealed that on the transport day, high concentrations of $PM_{2.5}$ occurred owing to the influence of long-range transport, while on the accumulation day, high concentrations of $PM_{2.5}$ occurred owing to the influence of atmospheric stagnation and accumulation. These findings indicate that different mechanisms contribute to $PM_{2.5}$ formation. The IPR analysis of the NF simulations showed that the transport and accumulation days were similar, with the EMIS (emission), AERO (aerosol process), and HADV (horizontal advection) processes contributing positively to the increase in $PM_{2.5}$, whereas ZTRA (vertical transport) and DDEP (dry deposition) contributed negatively. However, the YF simulation results showed changes in the physical/chemical processes due to the impact of the ADE. During transportation, the contribution of horizontal advection (HADV) increased as the $PM_{2.5}$ concentrations accumulated near the surface due to the ADE were transported from foreign countries to the Korean Peninsula. On the accumulation day, atmospheric stabilization (including decreased westerly winds and PBL heights) occurred owing to the ADE along with restrained vertical mixing. $PM_{2.5}$ concentrations increased due to atmospheric stabilization (reduced westerly winds and PBL heights), which reduced the outflow of horizontal advection (decreased HADV) and reduced the outflow of vertical transport (increased ZTRA) due to restrained vertical mixing. These resulted in an increase in $PM_{2.5}$ concentrations due to reduced inflow of horizontal advection (from 2.67 to 0.23 $\mu g \cdot m^{-3}$) and reduced outflow of vertical transport (from -4.34 to -3.38 $\mu g \cdot m^{-3}$).

When considering the ADE, stable atmospheric conditions and restrained vertical mixing can be significant factors contributing to the increase in $PM_{2.5}$ concentrations near the surface. However, the results of this study revealed that the physical and chemical factors leading to the increase in $PM_{2.5}$ varied depending on the $PM_{2.5}$. As a result, differences in the contributions from the processes were found between the transport and accumulation days. HADV and EMIS on the transport day and EMIS on the accumulation day were the main contributors to increasing $PM_{2.5}$ concentrations. Therefore, considering the ADE highlighted the contributions of processes related to $PM_{2.5}$ formation mechanisms.

This study investigated the ADE of meteorological conditions and air quality in South Korea, particularly examining the differences in impacts according to $PM_{2.5}$ formation mechanisms. However, a case study of air pollution in South Korea may not comprehensively elucidate the $PM_{2.5}$ formation mechanism. Also there is a technical limitation in not accounting for recent reduction pollutant emissions from abroad using historical overseas emission data, which could potentially result in an overestimation of the impact of long-range transport from foreign countries. In the future, updated overseas emission data should be utilized. However, it is important to note that the scientifically significant aspects of the findings in this study remain valid. Furthermore, synoptic patterns in relation to transport patterns from foreign countries into the Korean Peninsula and the horizontal and vertical distributions of $PM_{2.5}$ should also be investigated in future studies to understand their relationship with $PM_{2.5}$ formation mechanisms.

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