



A modeling study of PM_{2.5} transboundary transport during a winter severe haze episode in southern Yangtze River Delta, China

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ABSTRACT

The WRF-CHEM (Weather Research Forecast-Chemistry) model was used to investigate a severe haze episode in Zhejiang province in southern Yangtze River Delta, China, during 28 November to 10 December 2013 to obtain the characteristics of PM_{2.5} transboundary transport and its contributions to haze formation. The predicted temporal variations of PM_{2.5} and meteorological variables are in good agreement with in-situ observations in Zhejiang. Simulated wind direction and pollution levels in upwind areas have significant effects on modeled near-surface PM_{2.5} concentrations. Both the large-scale atmospheric circulations and local weather conditions contributed to the accumulation of pollutants in Zhejiang, featuring the weakened East Asia trough, the dominance of a weak high pressure system, a low wind speed and a low planetary boundary layer height (PBLH) during the heavily polluted period from 4 to 7 December. The near-surface PM_{2.5} concentration was highly positively correlated with the accumulated PM_{2.5} transport mass ($R = 0.88$ at a lag of 7 h) and moderately negatively correlated with the PBLH ($R = -0.39$) during this period, indicating that the horizontal transport across Zhejiang's boundary under stable meteorological condition played important roles in the high PM_{2.5} concentrations. The analysis of PM_{2.5} transboundary transport flux shows that the main transport pathway for outside pollutants affecting Zhejiang was mainly the northerly flow. There were strong input fluxes mainly within the planetary boundary layer on 4 and 6 December before the maximum concentration occurred in the early morning on 7 December. The contribution of PM_{2.5} transboundary transport to the aerosol pollution in the study domain was determined by a receptor-oriented model based on the conservation of mass formula for PM_{2.5}. The PM_{2.5} transboundary transport across the Zhejiang's boundary contributed 62% to the total PM_{2.5} increase during the severe haze episode, and the primary emission and secondary PM_{2.5} formation contributed 15% and 23%, respectively. This result suggests that the joint efforts with neighboring provinces to mitigate pollutant emissions are important to improve air quality in Zhejiang during winter.

1. Introduction

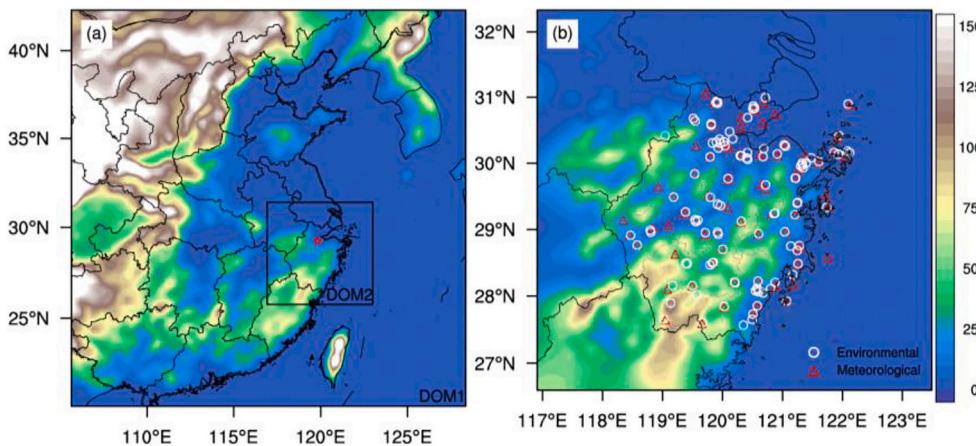
Haze has always been a “hot spot” concerning to all sectors of society since fine particulate matter (PM_{2.5}) poses very negative effects on human health, atmospheric visibility, and climate change (Hua et al., 2016; Ma et al., 2012; Zhang et al., 2012). The China Ecological Environment Bulletin in 2018 pointed out that the pollution events dominated by PM_{2.5} still accounted for the highest number of polluting days annually in the 169 key cities in China (P.R.C. MEE, 2019). The sources of PM_{2.5} consist of primary emissions and secondary transformation of gas precursors mainly from coal combustion, traffic sources, residential emission and natural events in China (Chan and Yao, 2008;

Wang and Hao, 2012; Wu et al., 2018). With rapid industrialization and urbanization in China, the emission intensities of anthropogenic activities vary among provinces/municipalities. The transport of PM_{2.5} originated from high emission regions affects the downwind areas under the prevailing wind and could be the major factor contributing to heavy haze pollution episodes (Jia et al., 2008; Wang et al., 2014a; Wang et al., 2018; Wu et al., 2017).

Zhejiang province is located in the south part of the Yangtze River Delta (YRD), of which the capital city is Hangzhou where the G20 summit 2016 was held. Besides, the 19th Asian Games 2022 will be hosted in this city. The plain and basin areas are distributed in the northern and central part of Zhejiang province, only accounting for

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23.2% of the total provincial region. Other areas are dominated by mountains and hills with the terrain slope from southwest to northeast (Fig. 1). The unique terrain open to the north facilitates the upwind pollution from northern adjacent provinces/municipality with higher emission intensities to be transported and accumulated in Zhejiang due to the mountain blockage when the weak northerly wind prevails in the YRD (Cao et al., 2011; Fu et al., 2016; Yu et al., 2017a; Yu et al., 2019). Furthermore, the emissions of air pollutants are increasing steadily with rapid economic development and high energy consumption in the YRD (Li et al., 2011; Niu et al., 2010; Pu et al., 2020; Sun et al., 2013; Xu et al., 2017). Therefore, it is necessary to distinguish the effects of transboundary transports and local emissions to have a better understanding of haze formation in Zhejiang.

The characteristics of the transboundary transport and its contribution to high aerosol pollution can always be investigated by computational atmospheric models because the continuous wind field data in atmosphere is essential (An et al., 2007; Chen et al., 2017; Lang et al., 2013; Li et al., 2015; Jiang et al., 2015; Sun et al., 2016; Wang et al., 2014b, 2016; Wang et al., 2018). For instance, Li et al. (2015) found that the southwest and south parts of Beijing were the source areas leading to high PM_{2.5} in Beijing during a period of fall in 2013 according to the results of hybrid receptor modeling. Wu et al. (2017) found out the non-Beijing emissions accounting for 61.5% of the PM_{2.5} level in Beijing during an episode of summer in 2015 based on the factor separation approach and WRF-CHEM model, which had the same variation trend as the PM_{2.5} horizontal transport flux across Beijing's boundary. Fu et al. (2016) have demonstrated that the slight or moderate pollutions were dominated by local contributions while regional transports significantly increased when the pollution aggravated in the YRD in the winter; besides, the residential emission was predicted as one of the major sources of PM_{2.5} derived from the zero-out method (Wu et al., 2018). There are some controversies about the role of non-local emissions in the air quality in Hangzhou (Gao et al., 2014; Fu et al., 2016; Wu et al., 2016; Yu et al., 2014). Local vehicle emission was suggested as a major contribution to the PM_{2.5} pollution in urban Hangzhou in the autumn of 2013 (Wu et al., 2016), and the sources affecting formation of the extremely high PM_{2.5} in Hangzhou were mainly located in the southeastern coast of Zhejiang and Fujian provinces, north part of Jiangxi, and central part of Jiangsu province during 3–9 December 2013 (Yu et al., 2014). Nevertheless, the trans-boundary transport characteristics of pollution from Shanghai and city clusters in southern Jiangsu province to Zhejiang have been neglected from the YRD regional perspective. The effect of mountain blockage to incoming pollution in Zhejiang cannot be taken into account when the Hangzhou city alone is considered. Based on our extensive literature review, there are very few studies which were focused on the roles of regional and local emissions of air pollutants in the heavy aerosol pollution in Zhejiang. Therefore, a thorough investigation about how

Fig. 1. Model domain configuration with elevation distribution (a) and locations of the meteorological and ambient monitoring sites (b). Red star denotes the location of Hangzhou city. Red triangles represent the meteorological sites and white circles indicate the ambient monitoring sites. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the transboundary transport would contribute to the regional haze episodes in Zhejiang is necessary, which might uncover more actionable insights to better inform more targeted emission control strategies in YRD region.

The impact of transboundary transport of pollutants across regions can be calculated by the zero-out method or the receptor models with the back-ward trajectory analysis. However, neither the zero-out method nor the receptor models can resolve the quantitative aerosol transport due to the complex pollutant sources outside the target region. In this study, a receptor-oriented method to quantify the contribution of PM_{2.5} transboundary transport to aerosol pollution is proposed based on the work from Jiang et al. (2015), which is developed according to the mass conservation of PM_{2.5} so as to effectively separate the effects of transboundary transport, local emission and secondary PM production. The objectives of this study are to: (1) analyze the relationship between aerosol pollution and synoptic pattern during a severe haze episode in Zhejiang province from the perspective of large-scale circulations and local weather conditions, based on surface and sounding observations, reanalysis data and WRF-CHEM simulation results; (2) investigate the impact of PM_{2.5} transboundary transport on high pollution levels by analyzing the temporal and spatial characteristics of PM_{2.5} horizontal transport flux; (3) quantify the contribution of PM_{2.5} transport to haze formation in Zhejiang by a receptor-oriented model based on conservation of mass formula for PM_{2.5}.

The paper is organized as follows: Section 2 describes the model configuration, observation data and methodology. Section 3 presents the validation of model results, the relationship between the near-surface PM_{2.5} concentration and synoptic pattern / PM_{2.5} transboundary transport, and then illustrates the contributions of PM_{2.5} transboundary transport to haze formation in Zhejiang province based on the receptor-oriented method, and discusses advantages and disadvantages of the method in details finally. In the end, major conclusions are summarized in Section 4.

2. Model and methodology

2.1. Model configuration

WRF-CHEM (version 3.6.1; Grell et al., 2005) was used in the simulation performed for the period from 28 November to 10 December 2013, because the haze event during this period represents the most severe pollution episode with the highest PM_{2.5} concentrations in Zhejiang province since the in-situ observation of PM_{2.5} started. A spin-up period of 40 h (00:00 UTC 26 November – 16:00 UTC 27 December 2013) was used to minimize the influence of the initial conditions and the output was obtained on an hourly basis. The simulation was conducted for central and eastern China with a 27-km horizontal grid spacing (Domain 1, Fig. 1a) and 36 sigma levels up to 50 hPa in the

vertical direction, using one-way nesting over Zhejiang province and surrounding areas at 9×9 km grid resolution (Domain 2, Fig. 1b). The meteorological initial and boundary conditions are based on the National Center for Environmental Prediction (NCEP) Final Analysis (FNL) reanalysis datasets with a grid resolution of $1^\circ \times 1^\circ$ and available every 6 h. The default profiles in WRF-CHEM were used as the chemical initial and boundary conditions for the domain 1, and those for the domain 2 are nested down from the output of domain 1.

Major physics options used include Morrison double-moment microphysics module (Morrison et al., 2009), the RRTMG shortwave and longwave radiation schemes (Iacono et al., 2008), the Yonsei University (YSU) Planetary Boundary Layer scheme (Hong et al., 2006), the Grell 3D ensemble cumulus parameterization (Grell and Dévényi, 2002) and the Noah land surface model (Chen and Dudhia, 2001). The Morrison double-moment microphysics scheme, RRTMG radiation schemes and the Grell 3D ensemble cumulus parameterization were implemented in WRF-CHEM to provide advanced treatments of relevant processes (Wang LT et al., 2016). The gas-phase chemistry mechanism chose Carbon-Bond Mechanism version Z (CBM-Z) (Zaveri and Peters, 1999) and the aerosol module was the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) using 4 sectional aerosol bins including aqueous reactions (Zaveri et al., 2008). The Fast-J photolysis scheme was applied for photolytic rate calculation (Wild et al., 2000). All of these physics and chemistry options selected in this work have been applied extensively in previous studies focusing on YRD (Liao et al., 2015; Sun et al., 2016; Wu et al., 2018; Yu et al., 2017b, 2019). The biogenic emissions were calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). The anthropogenic emissions were generated by the HTAP_v2 global emission grid map with a grid resolution of $0.1^\circ \times 0.1^\circ$ for the base year 2010 (Janssens-Maenhout et al., 2015), which used the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University including contributions from industry, power generation, agriculture, residential, and transportation sources of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCS), NH_3 , PM_{10} , $\text{PM}_{2.5}$, black carbon (BC), and organic carbon (OC) (He, 2012; Li et al., 2014). The MEIC inventory has been proved to provide reasonable estimations of emissions (Wang LT et al., 2014), and widely used in modeling studies about air pollution over eastern and northern China (Chen et al., 2017; Huang et al., 2014; Sun et al., 2016; Wang et al., 2016; Wu et al., 2018; Yu et al., 2017b).

2.2. Observation data

The meteorological data were obtained from the National Meteorological Information Center of China, including hourly air temperature at 2 m height, relative humidity at 2 m height, wind speed and wind direction at 10 m height of 71 stations in Zhejiang province. The hourly near-surface $\text{PM}_{2.5}$ concentration observation data of 108 ambient monitoring sites in Zhejiang province were acquired from the Ministry of Ecology and Environment of China. The locations of the meteorological and environmental sites are shown in Fig. 1b.

2.3. Methods

To quantify the relationship between the air pollution and the airflow transport, the $\text{PM}_{2.5}$ transboundary transport flux (\vec{F}) is defined as the horizontal wind field (\vec{U}) on the grid border multiplied by the $\text{PM}_{2.5}$ concentration of the corresponding grid (C) from which the airflow comes and then integrated on vertical layers as follows (Jiang et al., 2008; Wu et al., 2017b):

$$\vec{F}(t) = \int_{s=1}^N \int_{z=0}^H C(s, z, t) \cdot \vec{U}(s, z, t) \cdot \Delta x \, dz \, ds, \quad (1)$$

where N , H and Δx are the number of the boundary grids, the specified

height and the model horizontal grid spacing (9 km in this study), respectively. Most aerosol pollutants were centralized near the surface, while a small part of particles can also be transported to the height of 1–1.5 km to 2–3 km from the ground (Wang H et al., 2014). Therefore, this work focuses on the horizontal transport of $\text{PM}_{2.5}$ within a height of 3 km. The transport mass of $\text{PM}_{2.5}$ (M_{Trans}) can be achieved by integrating the transport flux defined in Eq. (1) in time.

According to the conservation of mass formula, the change of the total $\text{PM}_{2.5}$ suspended in the atmosphere in a fixed area with a high enough altitude (ΔM) should equal to the summation of the horizontal transport mass (M_{Trans}), the primary emission (M_{Emis}), the secondary formation (M_{Chem}) of $\text{PM}_{2.5}$ and the deposition (M_{Dep}) during a specified period:

$$\Delta M = M_{\text{Trans}} + M_{\text{Emis}} + M_{\text{Chem}} + M_{\text{Dep}}. \quad (2)$$

The total mass of $\text{PM}_{2.5}$ between the surface and the specified altitude was calculated by integrating $\text{PM}_{2.5}$ concentration of each model layer in both horizontal and vertical directions. The $\text{PM}_{2.5}$ primary emissions can be estimated from the anthropogenic emission inventory. M_{Dep} includes both dry and wet deposition and is always negative representing the removal of $\text{PM}_{2.5}$ in the vertical direction. In the WRF-CHEM model, not all the gas-phase chemical mechanisms and aerosol modules are coupled with the deposition diagnostics, so we modified the model so that the dry and wet deposition of $\text{PM}_{2.5}$ became the standard output variables. Finally, M_{Chem} can be obtained indirectly based on Eq. (2).

To further explore the contribution of $\text{PM}_{2.5}$ transboundary transport to the formation of haze, the contribution rate is defined as follows:

$$CR_T = \frac{M_{\text{Trans}}}{M_{\text{Trans}} + M_{\text{Emis}} + M_{\text{chem}}} \times 100. \quad (3)$$

And we can also calculate the contribution rate of local emission (CR_E) and secondary PM formation (CR_C) at the same time:

$$CR_E = \frac{M_{\text{Emis}}}{M_{\text{Trans}} + M_{\text{Emis}} + M_{\text{chem}}} \times 100, \quad (4)$$

$$CR_C = \frac{M_{\text{Chem}}}{M_{\text{Trans}} + M_{\text{Emis}} + M_{\text{chem}}} \times 100. \quad (5)$$

The contribution rate of $\text{PM}_{2.5}$ transboundary transport (CR_T) defined above denotes the contribution of transport of fine particles by air flow to the formation of haze. To achieve an effective CR_T , both M_{Trans} and ΔM need to be positive. Therefore, Eq. (3) adapts to any period with a significant increase of $\text{PM}_{2.5}$ concentration. The haze event during the study episode was the most severe in Zhejiang in recent years. The limitations and strengths of calculation method for the transboundary contribution rate will be discussed in details in Section 3.5.

3. Results and discussion

3.1. Model validation

Fig. 2 compares time series of simulated and observed meteorological variables averaged over Zhejiang province during the study period. The performance statistics are listed in Table 1 including the mean bias (MB), the root mean square error (RMSE), the normalized MB (NMB), the normalized mean error (NME) and the correlation coefficient (R) (Yu et al., 2006). The model showed good performance in predicting the temporal variations of air temperature at 2 m height (T2), relative humidity at 2 m height (RH2) and wind speed at 10 m height (WS10) with R values higher than 0.70, NMBs and NMEs ranging from -12% to 30% and from 16% to 37%, respectively. The wind direction at 10 m height (WD10) is moderately correlated ($R = 0.55$) with observations while exhibiting the smallest errors according to NMBs and NMEs among all tested meteorological variables. WS10 is overestimated to some extent, but the errors are lower than those in

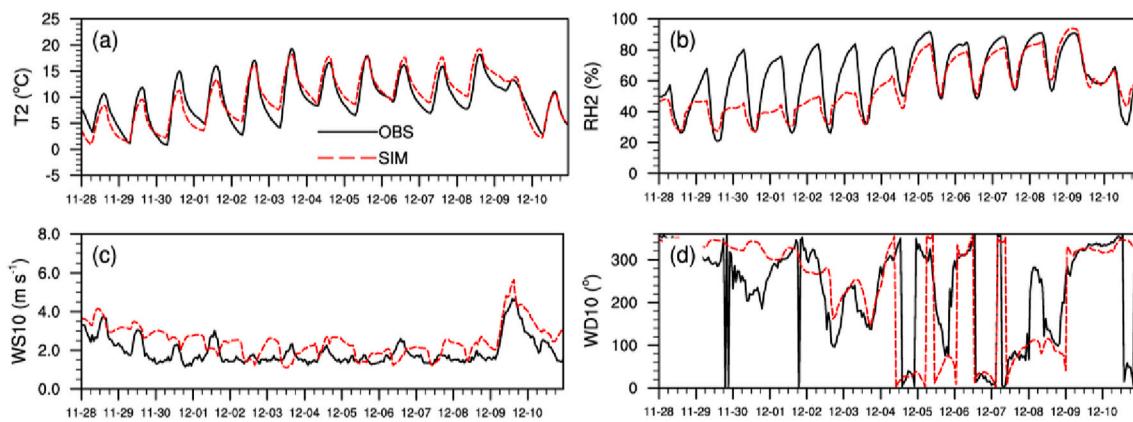


Fig. 2. Comparison of observed (black solid line) and simulated (red dash line) hourly air temperature at 2 m height (a), relative humidity at 2 m height (b), wind speed at 10 m height (c) and wind direction at 10 m height (d) from 28 November to 10 December 2013. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1

Performance statistics of simulated hourly air temperature at 2 m height (T2), relative humidity at 2 m height (RH2), wind speed at 10 m height (WS10), wind direction at 10 m height (WD10) and near-surface PM_{2.5} concentration in comparison with observations.

Variables	OBS	SIM	MB	RMSE	NMB(%)	NME(%)	R
T2 (°C)	9.6	9.9	0.3	1.9	3.5	17.2	0.91
RH2 (%)	63.2	56.0	-7.3	13.9	-11.5	16.0	0.78
WS10 (m s⁻¹)	1.9	2.5	0.6	0.8	29.7	36.7	0.72
WD10 (°)	325.4	338.5	8.2	57.0	2.5	12.3	0.55
PM _{2.5} (µg m⁻³)	136.9	116.1	-20.8	53.0	-15.2	28.1	0.77

previous studies (Liao and Liao, 2014; Tuccella et al., 2012; Wang et al., 2016; Zhang et al., 2010). Therefore, WRF-CHEM can reasonably capture the evolution of synoptic systems passing through Zhejiang province.

Comparison between the temporal variations of simulated and observed near-surface PM_{2.5} concentrations averaged over Zhejiang province is shown in Fig. 3a. The simulated PM_{2.5} concentrations are highly correlated ($R = 0.77$) with the observed values with the MB of $-20.8 \mu\text{g m}^{-3}$ mainly due to the underestimation for the period of 7–9 December, but the model captured the two peaks well during 3–8 December. The mean fractional bias (MFB) and mean fractional errors (MFE) (Yu et al., 2006) at each ambient monitoring site are exhibited in Fig. 3b and Fig. 3c, respectively. The model moderately overestimated the concentrations of PM_{2.5} in urban areas of Hangzhou, while slightly underestimated in most of the other areas. The MFBs range from -30% to 30% at most of sites (84%), and MFEs range from 25% to 50% at more than half of sites (65%) and are lower than 75% at the rest sites. Therefore, the model performance in reproducing near-surface PM_{2.5} concentrations in all sites varies from average ($\text{MFB} \leq \pm 60\%$ and $\text{MFE} \leq 75\%$) to good levels ($\text{MFB} \leq \pm 30\%$ and $\text{MFE} \leq 50\%$) (Morris et al., 2004; U.S. EPA, 2007).

A closer inspection indicates that large discrepancies of near-surface PM_{2.5} concentration during some periods are strongly related to the accuracy of the simulated wind field. Fig. 4 shows the relationship between the accuracy of the simulated wind direction and the model bias of PM_{2.5} concentration. The model generally underestimated PM_{2.5} concentration possibly due to the overestimated wind speeds and underestimated relative humidity compared with observations, which would affect the advection and hygroscopic growth of aerosols (Fig. 2b, c). Another possible reason could be the underestimation of the anthropogenic emission intensities (and/or total emissions) due to the economic development gap between the emission base year 2010 and the simulation period (November–December 2013). Nevertheless,

PM_{2.5} concentration was obviously overestimated during 28 November to 1 December 2013. The observed wind direction was westerly, but the simulated wind was biased to be north-northwesterly, and therefore, polluted air parcels from the north of Zhejiang province (seen in Fig. 9) were incorporated by the model leading to the overestimation of the PM_{2.5} concentration (Fig. 4b). The model significantly underestimated PM_{2.5} concentration during 7–9 December mainly due to the simulated sea breezes with higher frequencies and wind speeds than observations (Fig. 2c, 4d). Cleaner air parcels from the sea incorrectly reduced the aerosol pollution in Zhejiang province. During the haze episode from 2 to 6 December (Fig. 4c), the model showed a better performance in both wind direction and PM_{2.5} concentration compared with the other periods. Consequently, the relationship between PM_{2.5} concentration and wind speed was not linear, which was also influenced by wind direction and aerosol pollution levels in the upwind areas (Sun et al., 2016; Wang et al., 2018).

Another important factor influencing the accurate reproduction of the aerosol concentration is the prediction of the vertical temperature profile which is closely associated with the boundary layer height. Fig. 5 presents a comparison of the vertical temperature profiles obtained from sounding data and model results. Overall, the simulated profiles were well consistent with the observations, implying reasonable boundary layer height simulated by the model. But the model tended to overestimate the height of the temperature inversion on 5 December, showing that the actual stronger vertical diffusion was not well captured possibly leading to the overestimation of PM_{2.5} concentration in the early morning on 5 December (Fig. 3a, 4c, 5c). On the contrary, the simulated temperature inversion height was much lower than the observation on 7 December, indicating that the actual more stable weather was not well captured resulting in the underestimation of aerosol concentration under more frequent sea breezes by the model (Fig. 3a, 4d, 5d).

3.2. Relationship between aerosol pollution and synoptic pattern

Fig. 6 depicts the spatial distributions at 500 hPa geopotential height and 850 hPa wind at 08:00 BJT over East Asia during the study episode based on NCEP-FNL reanalysis data. The strong East Asia trough located in eastern China brought dry and cold air to Zhejiang from high latitudes on 28 November (Fig. 6a). The trough weakened and moved to northeast part accompanying with the gradual sparse isobaric lines over Zhejiang, suggesting the weakness of the northwesterly winds from 28 November to 1 December (Fig. 6b, c). Subsequently, most areas of Zhejiang were mainly dominated by weakened pressure field gradient implying stagnant air conditions during the severe pollution process from 4 to 7 December (Fig. 6d–6g), which is

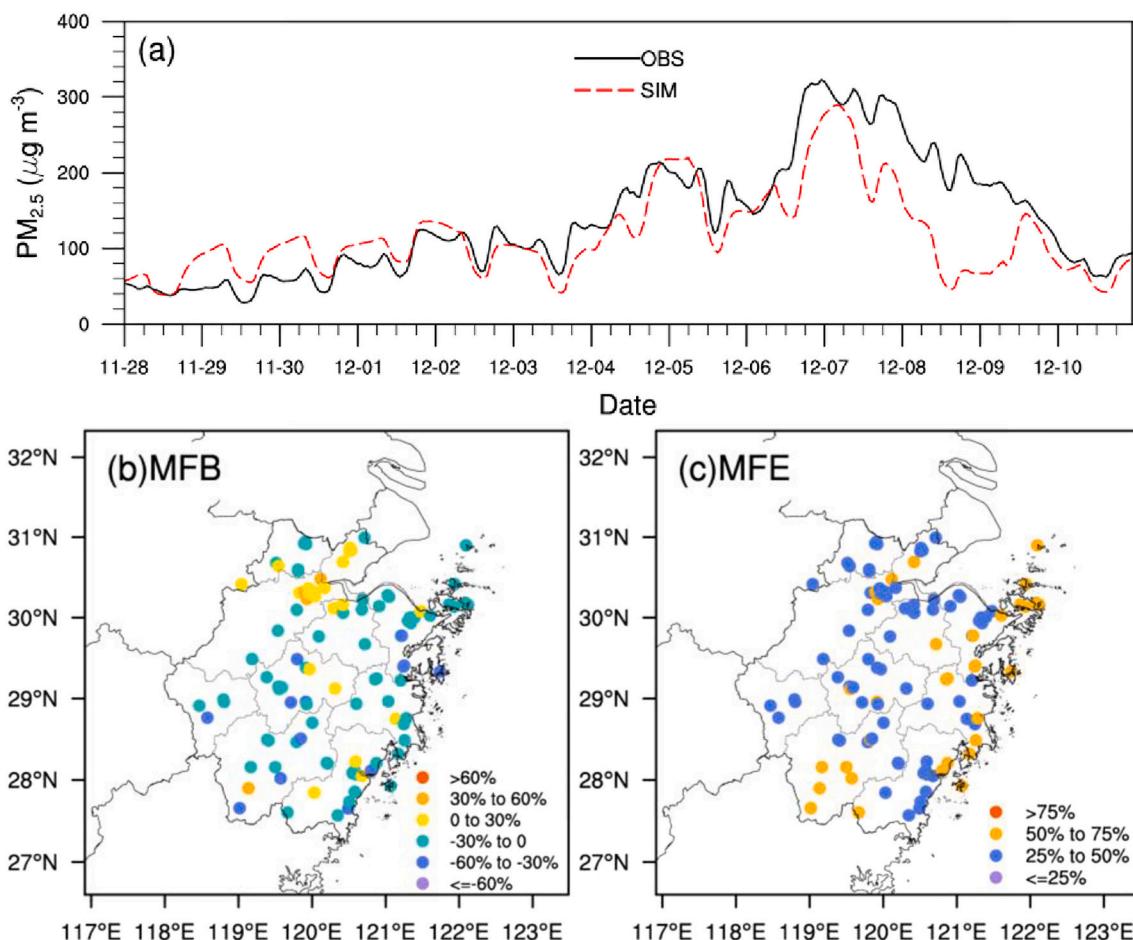


Fig. 3. Comparison of the simulated (red dash line) and observed (black solid line) hourly near-surface PM_{2.5} concentration, temporal variations of domain-wide mean (a), and spatial distribution of MFB (b) and MFE (c).

unfavorable for horizontal dispersion of pollutants in Zhejiang. The mid-latitude trough was still weak and the associated northerly wind extended to the north rather than south of Zhejiang during this period favorable for the transport of pollutants from outside to Zhejiang and accumulation. 850 hPa wind fields showed a weak high pressure system over Zhejiang was not conducive to the vertical diffusion of aerosols from 5 to 7 December. Additionally, warm and humid airflows from the East China Sea promoted the haze formation and maintenance. After that, the deepened trough enhanced the dry and cold northwesterly flows to Zhejiang and increased the wind speed during 9–10 December (Fig. 6h, i), which was beneficial to horizontal dispersion of aerosols so as to alleviate the PM_{2.5} pollution episode in Zhejiang.

Statistics of the hourly simulated near-surface winds averaged over Zhejiang province are illustrated by wind rose plots in Fig. 7. Strong northerly and northwesterly winds ($> 3 \text{ m s}^{-1}$) of high frequency dominated in Zhejiang during the clean and slightly polluting periods with the averaged wind speed of 3.3 and 4.2 m s^{-1} , respectively (Fig. 7a, d). This is beneficial for air pollutants to be transported out from Zhejiang. The heterogeneous wind directions and low wind speed suppressed the advection of pollutants during 1–6 December when the haze began to form (Fig. 7b). The weak easterly and southeasterly humid flows of high frequency facilitated the particle formation and hygroscopic growth to worsen the aerosol pollution on 7 December with a strong PM_{2.5} concentration peak (Fig. 3a). After that, the clean air from the East China Sea helped alleviate the pollution levels leading to the decline of PM_{2.5} concentrations subsequently.

Fig. 8 (a) shows the simulated altitude-time cross-sections of PM_{2.5} concentrations and the planetary boundary layer height (PBLH)

averaged over Zhejiang. It can be seen that most particulate matter was suspending below 1500 m height. The concentration level below 300 m height was very high from 4 to 7 December, especially during the nighttime, attributable to the lower PBLH from 20:00 BJT to 08:00 BJT and lower wind speed (Fig. 7), which led to the weak convection and advection of atmospheric pollutants. During this period, PM_{2.5} concentration near the surface exceeded 150 $\mu\text{g m}^{-3}$ for 42 h accounting for almost half of the haziest period and it was still higher than 120 $\mu\text{g m}^{-3}$ at 500 m altitude till the early morning of 7 December. In addition, the PM_{2.5} concentration was consistently high ($> 120 \mu\text{g m}^{-3}$) from the ground to the altitude at 500 m in the afternoon on 9 December though the PBLH remained high due to strong convection in the afternoon. Furthermore, PBLHs during the nighttime from 1 to 8 December, when the haze started to form and maintained, were significantly lower than the other nights.

3.3. Characteristics of transboundary transport of PM_{2.5} and its relationship with the near-surface PM_{2.5} concentration

To further explore the relationship between near-surface PM_{2.5} concentration and the airflow transport, the accumulated PM_{2.5} transport mass (AM) is defined as the integration of net transboundary transport flux below 3 km height from the initial time of the analysis (00:00 BJT 28 November) to each time along Zhejiang's boundary. The temporal variations of the modeled PM_{2.5} concentration and the AM are presented in Fig. 8b. The near-surface concentration of PM_{2.5} showed two spikes right after the two peaks of AM occurred during 4–7 December with R of 0.54 and at a lag of 7 h up to 0.88 (the maximum lag

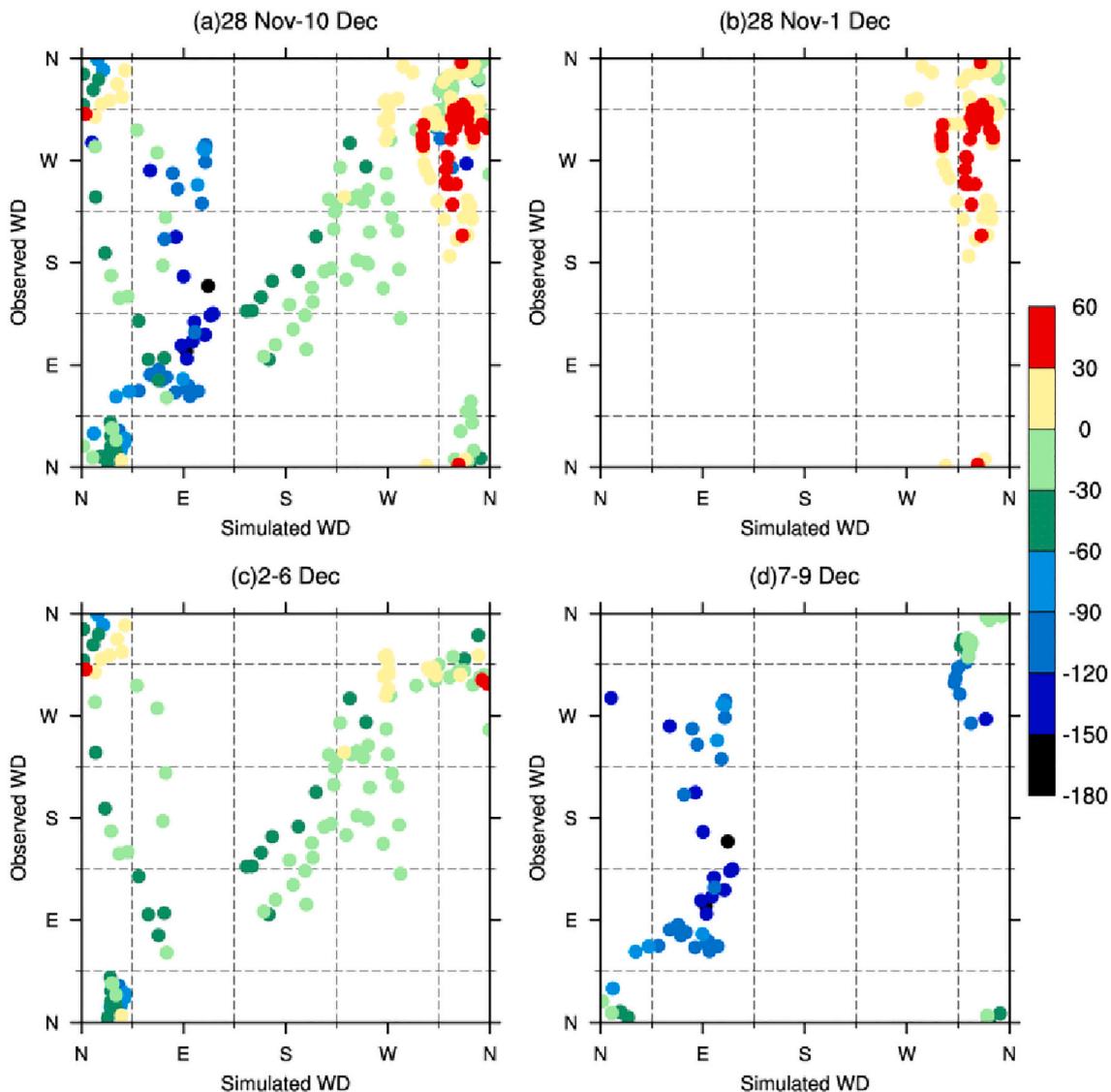


Fig. 4. The relationship among observed wind direction at 10 m height, simulated wind direction at 10 m height, and bias in modeled .vs. observed PM_{2.5} concentration (colored dots) from 28 November to 10 December (a), from 28 November to 1 December (b), from 2 to 6 December (c), and from 7 to 9 December (d). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

correlation coefficient), concurrently corresponding to minimum PBLH at night with R of -0.39 during this period. This implies that the strong regional transport of pollutants and weak vertical diffusion played important roles in the extremely high aerosol concentrations within Zhejiang. In addition, the peaks of PM_{2.5} concentration and the AM occurred almost at the same time in the afternoon of 9 December, when however the relatively high PBLH was observed favorable for the vertical diffusion of pollutants. This led to highly positive correlations between the PM_{2.5} concentration and AM / PBLH on that day with R values of 0.91 and 0.86, respectively. Therefore, the PM_{2.5} trans-boundary transport may be the key factor to build up the peak of PM_{2.5} concentration on 9 December.

From the spatial distribution chart of daily PM_{2.5} transport fluxes and near-surface concentrations (Fig. 9), it can be seen that the haze formed in Jiangsu and Anhui provinces on the north of Zhejiang firstly, strengthened and then extended to Zhejiang under the prevailing northerly winds from 30 November to 6 December (Fig. 9b-9f). Before 1 December, strong northerly wind ($> 3 \text{ m s}^{-1}$) prevailed in the whole study domain and the PM_{2.5} concentration was relatively low (Fig. 7a, 9a, b). Subsequently, the northerly wind weakened ($< 2 \text{ m s}^{-1}$) and the air quality rapidly deteriorated due to the strong particle fluxes

transported from the north and northwest polluted areas in Jiangsu and Anhui to Zhejiang on 1, 4 and 6 December (Fig. 7b, 9c-9e). The PM_{2.5} transport flux was weak in southern Zhejiang due to the relatively low wind speed and pollution level, favorable for pollutant accumulation over middle and northern Zhejiang. Hence, the graphical coverage and levels of aerosol pollution gradually increased in Zhejiang until 7 December, which is well consistent with the high lag correlation between PM_{2.5} concentration and the accumulated transport mass of fine particles (AM). The transport flux turned from northerly to southerly on 7 December, the northern Zhejiang became severely polluted area, where the daily PM_{2.5} concentration exceeded $300 \mu\text{g m}^{-3}$ (Fig. 9f). Meanwhile, the PM_{2.5} concentration averaged over Zhejiang reach the maximum value of $244.9 \mu\text{g m}^{-3}$ at 03:00 BJT on that day. Strong northwest wind ($> 4 \text{ m s}^{-1}$) was prevailing and the PM_{2.5} concentration declined to relatively low levels over the north of Zhejiang on 9 December (Fig. 9g). The largest flux intensities due to high wind speeds covered the whole study domain presenting very strong pollutant input from and output across the northwest and southeast borders of Zhejiang, respectively. Eventually, the PM_{2.5} concentration dropped to below $70 \mu\text{g m}^{-3}$ on 10 December (Fig. 9h).

Therefore, the main transport pathways for the extra-provincial

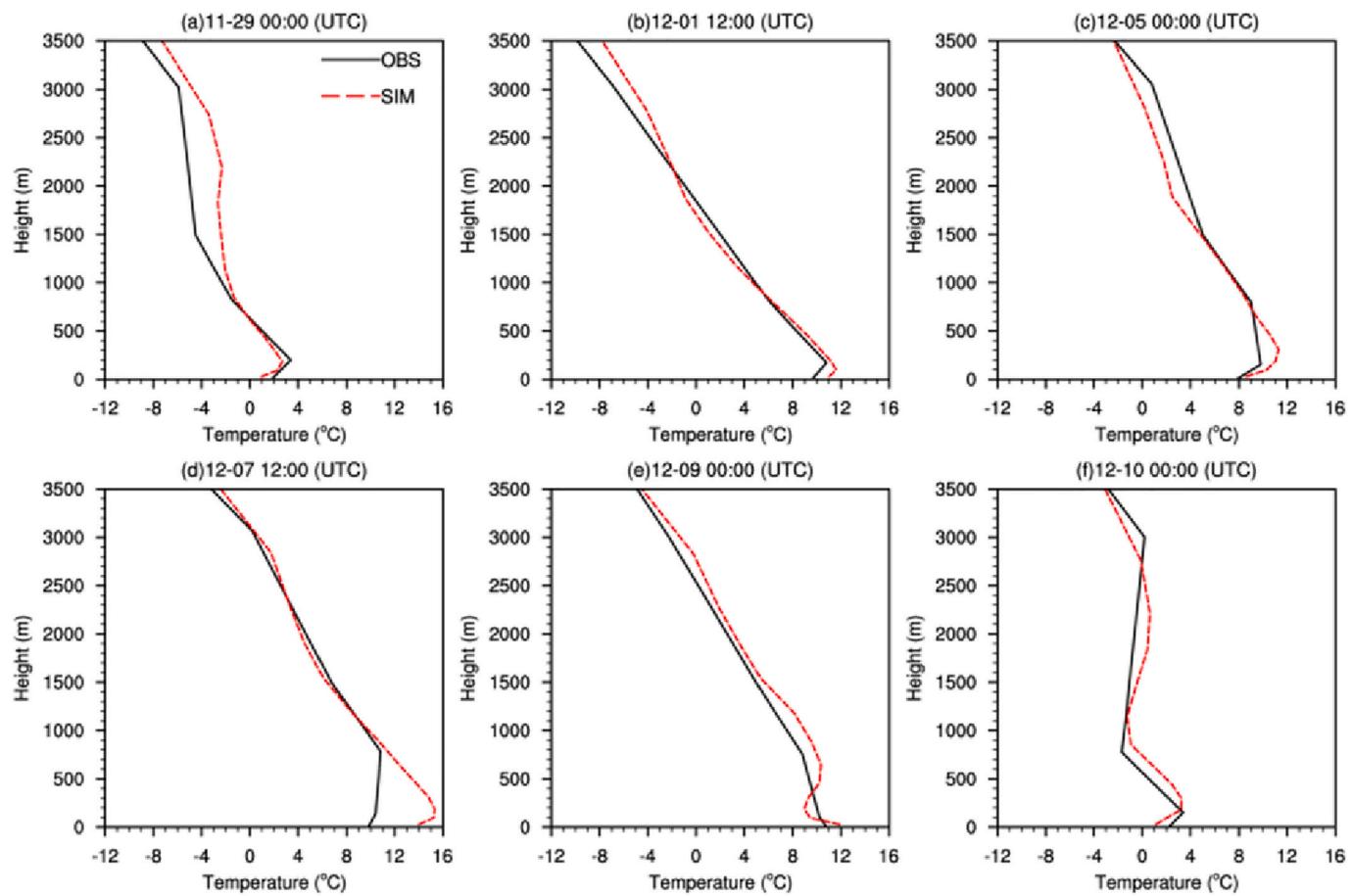


Fig. 5. Comparison of simulated and observed vertical temperature profiles for the Hangzhou (58457) site. The sounding data were achieved from: <http://www.weather.uwyo.edu/upperair/sounding.html>.

pollutants affecting Zhejiang were the northerly and northwesterly flows during this episode. The synoptic pattern shows that Zhejiang was dominated by weak northerly wind on 850 hPa and quite sparse 500 hPa isobars due to the weakened mid-latitude trough during 4–6 December (Fig. 6), which is very conductive for the extra-provincial pollutants in north region on Zhejiang to be transported into Zhejiang.

The daily net horizontal transport flux of $\text{PM}_{2.5}$ across the boundary of Zhejiang province and the vertical profiles of the flux on the selected periods are displayed in Fig. 10 and Fig. 11. As shown in Fig. 10a, the net fluxes below 3 km altitude were positive on 1, 4, 6 and 9 December with the values of 23.5, 66.9, 66.6 and 28.6 kg s^{-1} , respectively, suggesting that $\text{PM}_{2.5}$ was transported to Zhejiang and mainly from the north and west as shown in Fig. 10b. The positive net fluxes within the planetary boundary layer (PBL) only occurred on 4 and 6 December (because the positive flux was close to zero on 3 December) mainly from the north and accounted for 60% and 75% of those below 3 km height, respectively, indicating that transboundary transport mainly occurred within the PBL under the circumstance of strong input of pollutants. This feature is also seen in the vertical distribution of the transport flux. The positive fluxes were primarily below the mean altitude of about 1000 m on 4 and 6 December and the fluxes below 500 m height were much stronger on 6 December with the highest value of about 20 kg s^{-1} near the altitude of 250 m (Fig. 11b, c). While the fluxes were positive mainly between the altitude of 500 m and 2000 m on 1 and 9 December, the peak values occurred above 1000 m height (Fig. 11a, f). Therefore, the positive fluxes below 3 km height on 4 and 6 December, mostly occurring in the boundary layer, played an important role in the high $\text{PM}_{2.5}$ concentrations in the early morning on 5 and 7 December. Zhejiang acted as a strong pollutant sink on 4 and 6

December, receiving $\text{PM}_{2.5}$ from its surrounding regions.

The maximum output fluxes below 3 km height are found on 7 and 8 December mainly from the north with a value of -130.2 and -95.6 kg s^{-1} , respectively, due to the prevailing easterly and southeasterly winds (Fig. 7c). But the negative fluxes within the PBL were much smaller than below 3 km height, especially on 7 December the flux was close to zero, implying that large pollutant transport out from Zhejiang occurred above the PBL. The PBL heights have significant differences on land and at sea in different seasons because of sea-land temperature differences, reflecting in higher PBLs at sea than on land in autumn and winter, and vice versa in spring and summer. On 7 December, the $\text{PM}_{2.5}$ concentrations were relatively low over the coastal areas compared with inland areas. Therefore, the inflow of $\text{PM}_{2.5}$ from east boundary of Zhejiang offset the outflow from the west boundary under the prevailing easterly wind within the PBL (Fig. 10b). Meanwhile, high humidity ($> 80\%$) accompanied with low PBLH in the early morning and evening, which facilitated particle transformation and hygroscopic growth. Consequently, the maximum value of $\text{PM}_{2.5}$ concentration appeared on 7 December as a result of the synergistic effects of transported-in pollutant accumulation, particle transformation and continuous local emissions.

The vertical profiles of $\text{PM}_{2.5}$ transport flux on 7 and 8 December generally show similar patterns with strong negative net fluxes below 500 m altitude (Fig. 11d, e). A large quantity of $\text{PM}_{2.5}$ transported out from Zhejiang mainly above the PBL would aggravate the pollution levels in surrounding regions. Accordingly, Zhejiang served as a strong pollutant source on 7 and 8 December transporting $\text{PM}_{2.5}$ out to surrounding areas after the most severe haze episode.

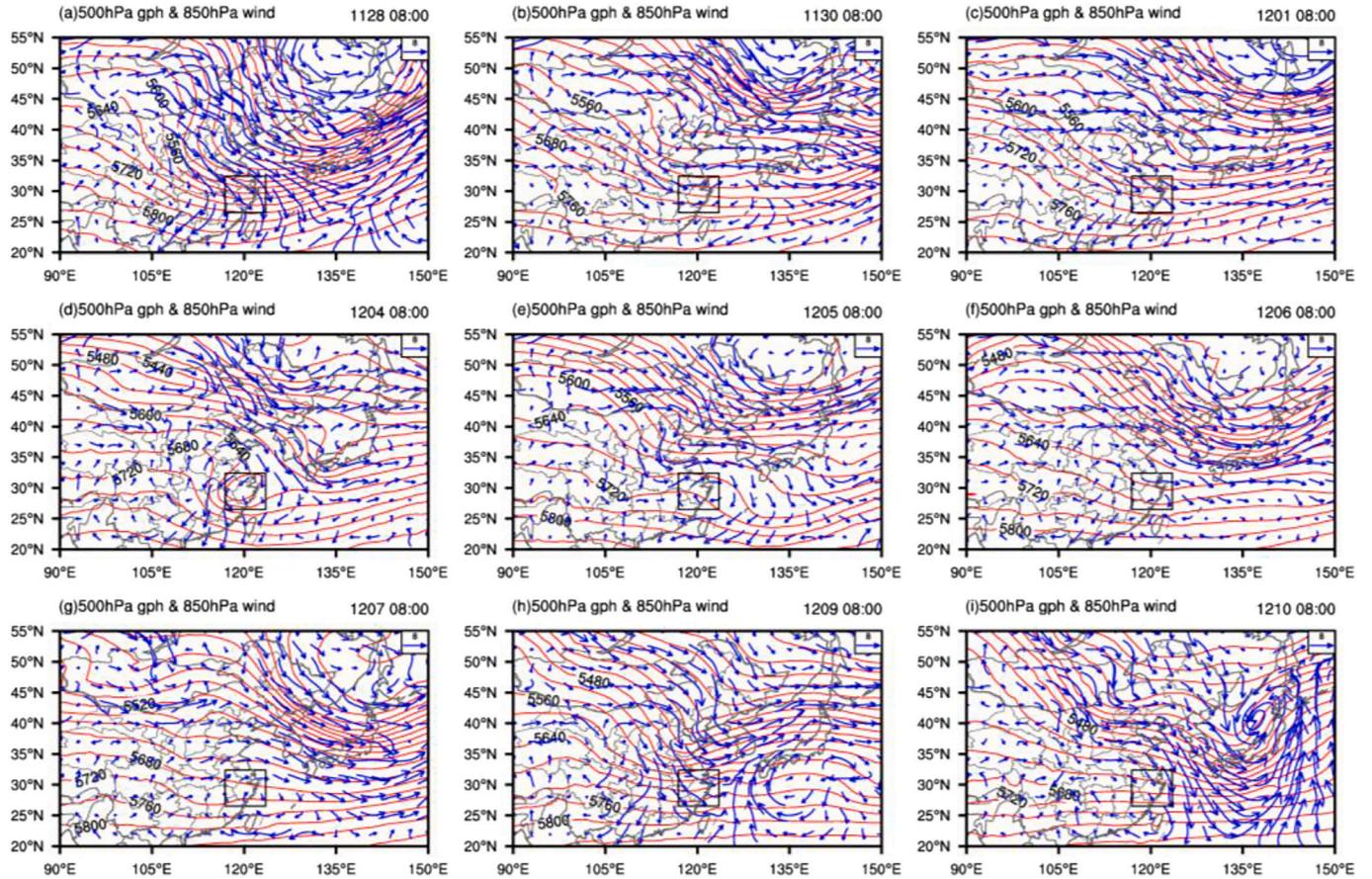


Fig. 6. 500 hPa geopotential height (m) and 850 hPa wind field (m s^{-1}) at 08:00 BJT over East Asia from 28 November to 10 December 2013.

3.4. $\text{PM}_{2.5}$ transboundary transport contribution to haze formation

The analysis above has demonstrated the strong correlation between the $\text{PM}_{2.5}$ transboundary transport and the high pollution level in Zhejiang during the study episode. Besides, it is essential to confirm the contribution of $\text{PM}_{2.5}$ transboundary transport on haze formation. Fig. 12 exhibits the temporal variations of the transboundary transport flux of $\text{PM}_{2.5}$, the total $\text{PM}_{2.5}$ suspended below 3 km height and the near-surface $\text{PM}_{2.5}$ concentration. Table 2 lists components in the conservation of mass formula for $\text{PM}_{2.5}$ (Eq. (2)) during two different periods. The hourly total mass and near-surface concentration of $\text{PM}_{2.5}$ showed the similar variation trend with the correlation coefficient up to 0.90 during the study period due to most particles accumulated in the near ground atmosphere. Thus high levels of the total mass of $\text{PM}_{2.5}$ can reflect high aerosol concentrations to a large extent.

The total mass of $\text{PM}_{2.5}$ below 3 km height in Zhejiang significantly rose from the valley value of 4266.5 ton at 11:00 BJT on 3 December to the maximum value of 21,378.1 ton at 21:00 BJT on 6 December corresponding to wavy increase in the near-surface $\text{PM}_{2.5}$ concentration, which is a typical case for the application of the receptor-oriented model based on conservation of mass formula to calculate the contribution of $\text{PM}_{2.5}$ transboundary transport to haze formation (Eq. (2–5)). During this period, the total mass of $\text{PM}_{2.5}$ increased by 17,111.6 ton and the transport mass were 10,945.6 ton by the summation of the transboundary transport flux over time. Large quantities of $\text{PM}_{2.5}$ input especially within the PBL from outside of Zhejiang and accumulated under the dominant weak northerly wind, because the aerosol pollution first occurred in regions on the north of Zhejiang, e.g. Jiangsu province, Anhui province and Shanghai (Fig. 9). According to Eq. (3), the transboundary transport mass of $\text{PM}_{2.5}$ accounted for 62% of the total mass increase from 11:00 BJT on 3 December to 21:00 BJT

on 6 December, which contributed most to the formation of haze. The contributions of the primary emission and local secondary formation of $\text{PM}_{2.5}$ were 15% and 23%, respectively (Eq. (4–5)).

In addition, the total mass and near-surface concentration of $\text{PM}_{2.5}$ rapidly rose together from their valley values at 01:00 BJT on 9 December and reached the peak simultaneously at 14:00 BJT on 9 December with strong input flux intensities though the PBLH were relatively high due to strong convection in the afternoon. The transport mass of $\text{PM}_{2.5}$ (17,992.7 ton) exceeded the increase of the total mass (16,899.4 ton) due to the wet deposition caused by precipitation in the morning of 9 December. The contribution of fine particle pollutants from surrounding areas during this polluted period was up to 93% based on Eq. (3), and the local inputs including emission and particle formation only accounted for 7%.

3.5. Advantages and disadvantages of the receptor-oriented method

The method based on conservation of mass formula for $\text{PM}_{2.5}$ is available for any period with a significant increase in near-surface $\text{PM}_{2.5}$ concentration and total mass of $\text{PM}_{2.5}$, which exactly quantifies the proportion of $\text{PM}_{2.5}$ accumulation contributed by the air flow transport to the $\text{PM}_{2.5}$ level elevation. The contribution rate estimated by the receptor-oriented model is supposed to be part of the contribution from external pollutant sources due to two limitations. On the one hand, the local secondary $\text{PM}_{2.5}$ formation in the formula includes chemical transformation of gas precursors transported from outside of the selected region. On the other hand, the $\text{PM}_{2.5}$ output from the selected region contains both transport-derived and local-derived fine particles. When both the secondary $\text{PM}_{2.5}$ production derived from external gas precursors and the local-derived $\text{PM}_{2.5}$ output are relatively small, the contribution of $\text{PM}_{2.5}$ transboundary transport to the

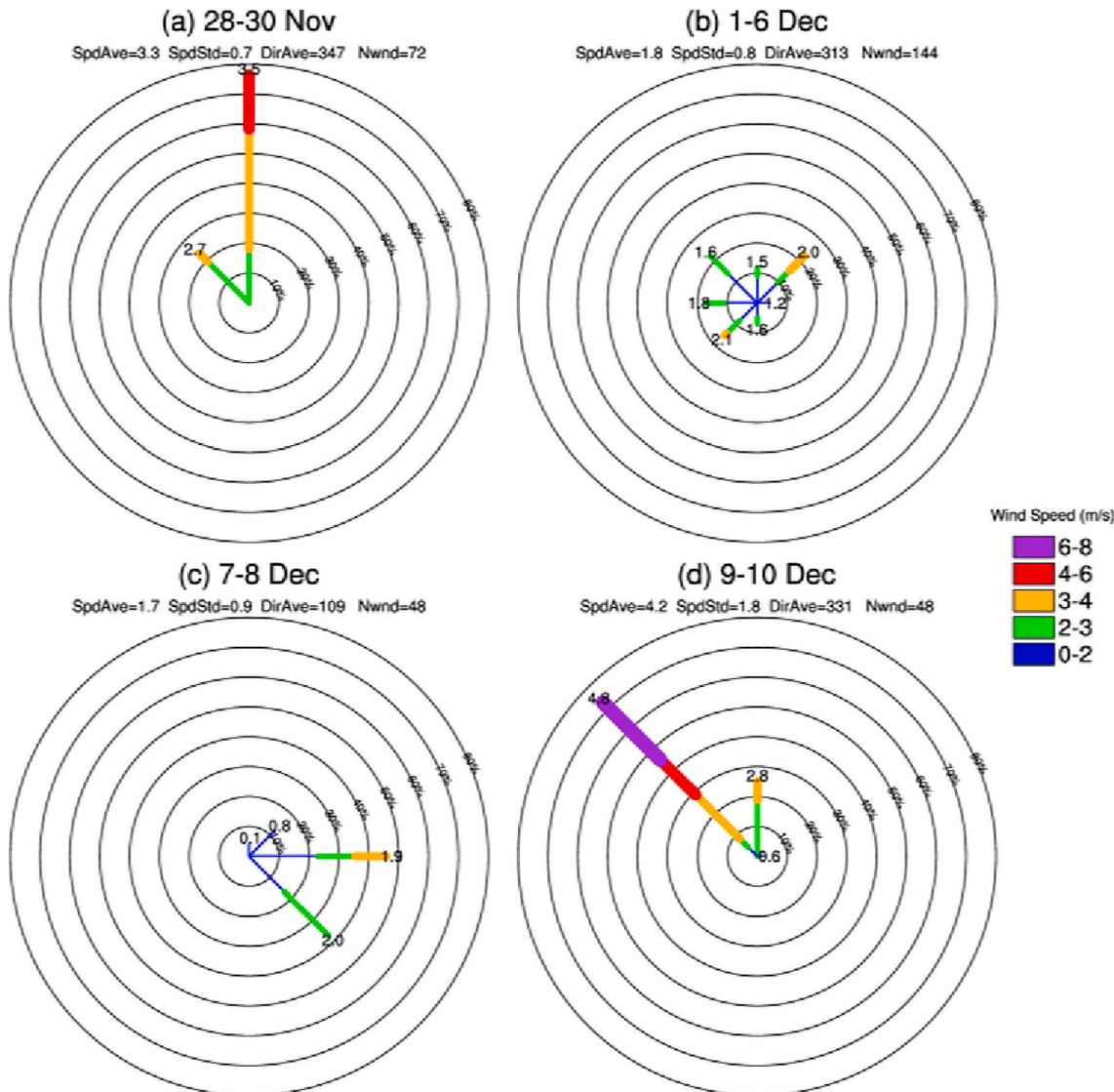


Fig. 7. Wind rose plots of near-surface wind field averaged over Zhejiang during four phases from 28 November to 10 December 2013. Four phases are (a) 28–30 November, (b) 1–6 December, (c) 7–8 December and (d) 9–10 December, respectively.

total PM_{2.5} increase is approximately equal to the contribution of external pollutant sources to the PM_{2.5} level. The advantage of the method is that efficient computation based on only one numerical simulation without auxiliary complex models is suitable for operational forecasting to predict the impact of PM_{2.5} transboundary transport on haze formation. This approach was originated from the study of Jiang et al. (2015), which has revealed that the transport of PM_{2.5} from Beijing's environs contributed about 55% to total PM_{2.5} increases during a severe haze episode in December 2013 using a rough square boundary surrounding Beijing and even ignoring the deposition of PM_{2.5}. It should be noted that the deposition of aerosols especially the wet deposition should be considered in the humid regions such as Zhejiang province to ensure the correct calculation of the contribution rate in Eq. (3). If the wet deposition is neglected, the contribution rate may exceed 100% which is unreasonable (Table 2).

There are a few studies to investigate the impact of the transboundary transport on haze in Hangzhou city but few works focusing on Zhejiang province. Yu et al. (2014) have demonstrated that air mass pathways and cross-border transport controlled high PM_{2.5} concentrations and formation in Hangzhou during 3–9 December 2013. Gao et al. (2014) have showed that non-Hangzhou pollutants made significant contributions up to 70% to the PM_{2.5} pollution in Hangzhou during a

heavy haze episode in winter in 2011, and the contribution of non-Zhejiang pollutants accounted for about 60% (Fu et al., 2016). To further validate the results in this work, another sensitive test was conducted by the zero-out method eliminating emissions in Zhejiang province during the same episode. The results showed that the contribution of pollutant sources outside Zhejiang to PM_{2.5} concentration ranged from 69.8% ~ 76.1% during the severe haze period with the near-surface PM_{2.5} concentration beyond 200 $\mu\text{g m}^{-3}$ during 6–7 December and ranged from 88.5% ~ 95.3% near the PM_{2.5} peak on 9 December. This is consistent with the results shown in Table 2 implying a leading role of external pollutant transport especially particle transport in the formation and maintenance of haze.

It depends on a number of factors, especially the time scale of the study period and the local emission characteristics, whether the transboundary transport plays a dominant role in the air quality. Generally speaking, transboundary transport may be primarily responsible for the severe haze events during several days (Chen et al., 2017; Jiang et al., 2015; Lang et al., 2013; Sun et al., 2014; Tao et al., 2014; Wang et al., 2014c; Yu et al., 2014), but the local emission or secondary PM production probably contributes most on a larger time scale such as monthly or annually (Wang et al., 2014a; Wang et al., 2015, 2018; Wu et al., 2016; Wu et al., 2018). However, combined with typical traffic

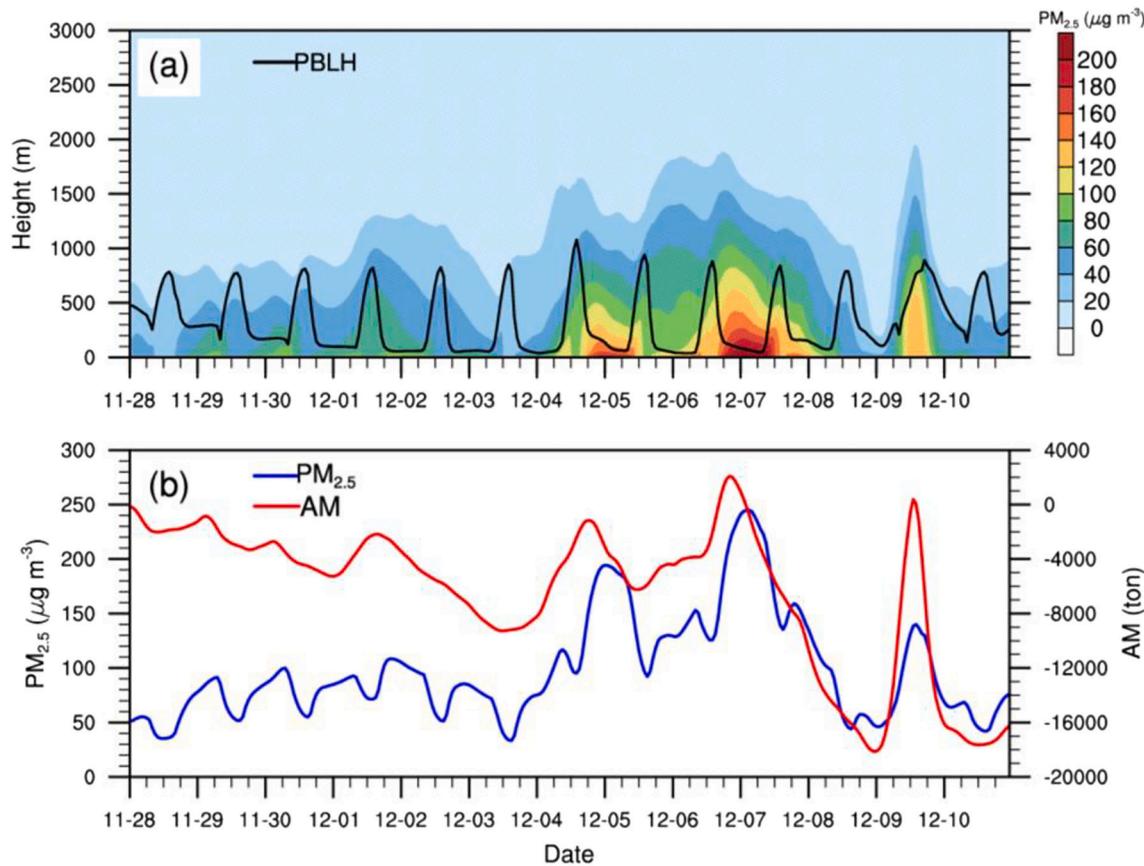


Fig. 8. Vertical distribution of the average PM_{2.5} concentrations and the planetary boundary layer height (PBLH) in Zhejiang (a) and the temporal variations of near-surface PM_{2.5} concentration and the accumulated PM_{2.5} transport mass (AM) (b).

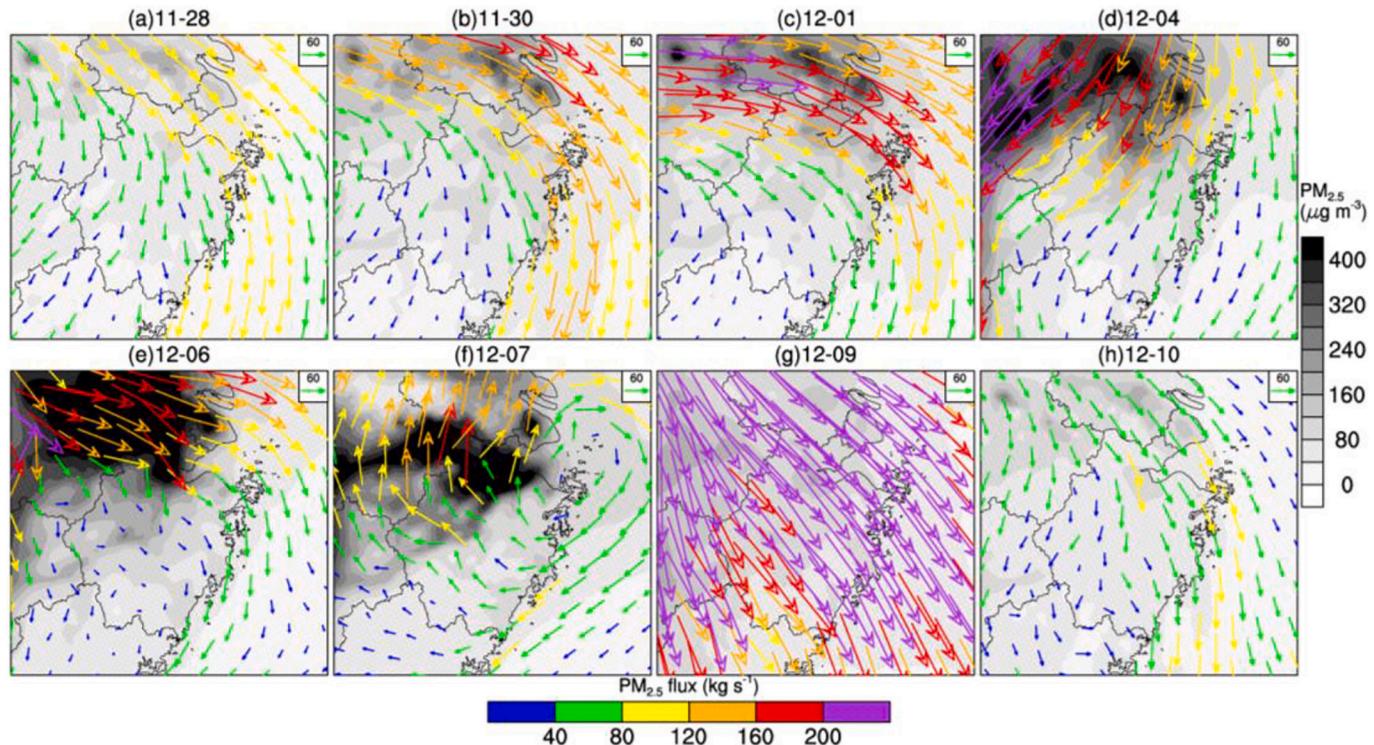


Fig. 9. Spatial distributions of daily PM_{2.5} horizontal transport flux below 3 km altitude (colored arrows) and the near-surface PM_{2.5} concentration (grey contour) from 28 November to 10 December 2013. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

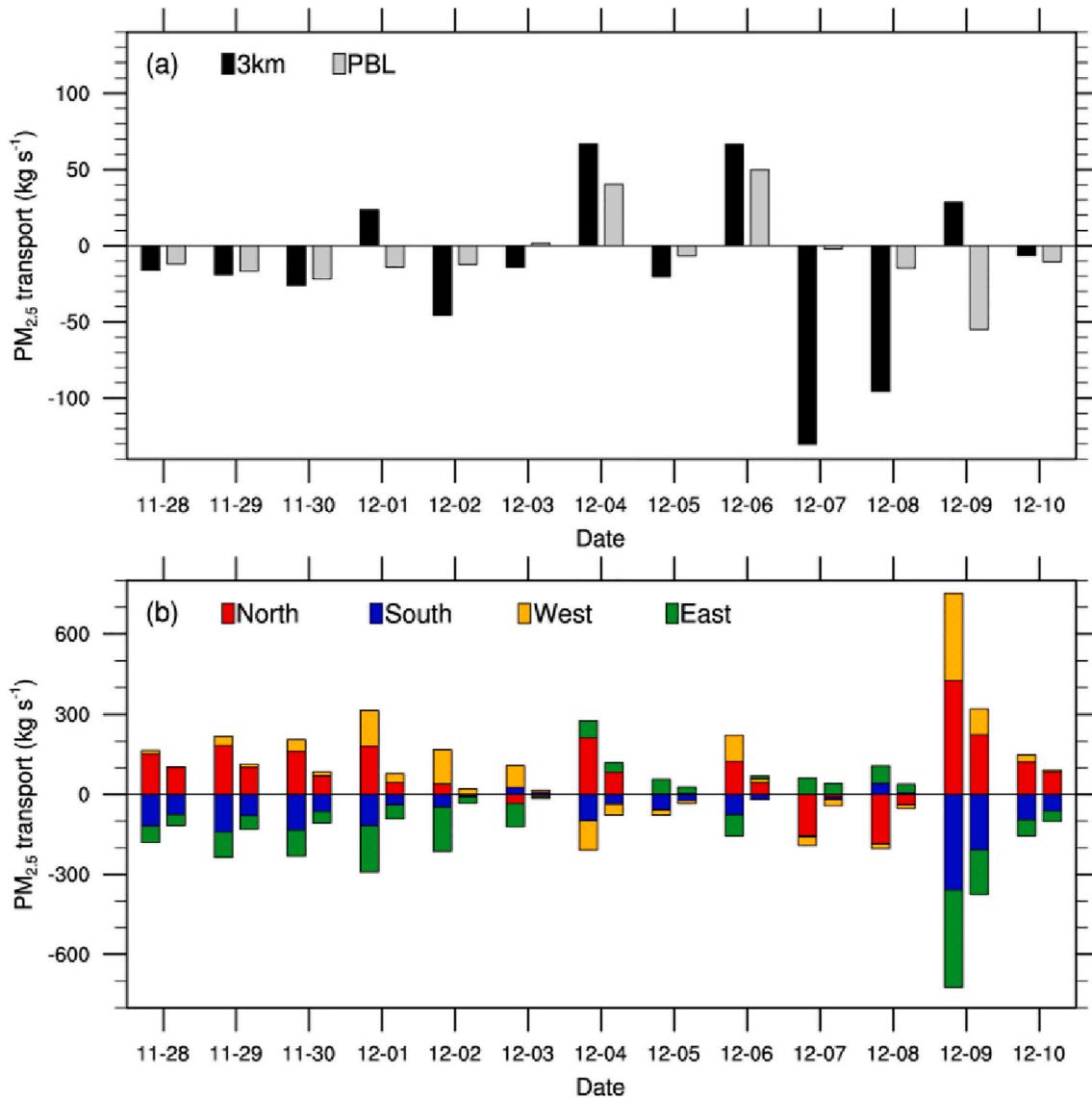


Fig. 10. Daily net transboundary transport flux of $\text{PM}_{2.5}$ below 3 km height (left column) and within the planetary boundary layer (PBL) (right column) integrating along the boundary of Zhejiang (a) and along the north, south, west, east boundaries of Zhejiang (b).

emissions on the urban scale favorable for efficient secondary PM formation by photochemical oxidation of VOCs and NO_x , this issue may become more complicated and cannot come to an agreement for high $\text{PM}_{2.5}$ level (Guo et al., 2014; Li et al., 2015; Wu et al., 2016; Zhang et al., 2015). The $\text{PM}_{2.5}$ transboundary transport played a major role in haze formation during 28 November to 10 December 2013 in Zhejiang province by means of a receptor-oriented model based on conservation of mass formula for $\text{PM}_{2.5}$ in this study, which is further proved by zero-out method. A comprehensive analysis taking advantage of dispersion modeling, receptor modeling and in-situ measurements for the secondary aerosol constituents on different time and spatial scales would exhaustively elucidate the contributions of transboundary transport to the air quality in Zhejiang province.

4. Conclusion

A winter severe haze episode with an extremely high concentration of $\text{PM}_{2.5}$ in Zhejiang province is simulated using the WRF-CHEM model during the period from 28 November to 10 December 2013, to evaluate the characteristics and contributions of $\text{PM}_{2.5}$ transboundary transport to the formation and maintenance of haze in Zhejiang province.

Adverse weather conditions played an important role in the high $\text{PM}_{2.5}$ concentrations. The weakened East Asia trough decreased the pressure field gradient in Zhejiang and blocked the northerly wind from running through Zhejiang accompanied by air mass subsidence under the control of a weak high pressure system at 850 hPa, suppressing the convection. Weak near-surface wind speed ($< 2 \text{ m s}^{-1}$) and low PBLH trapped pollutants during heavily polluted days. Particle formation and hygroscopic growth further aggravated the pollution due to humid air flow from the East China Sea by the easterly wind on 7 December leading to the occurrence of the highest $\text{PM}_{2.5}$ concentration. The near-surface $\text{PM}_{2.5}$ concentration was highly positively correlated with the accumulated $\text{PM}_{2.5}$ transport mass (AM) ($R = 0.88$ at a lag of 7 h) and moderately negatively correlated with the PBLH ($R = -0.39$) when the $\text{PM}_{2.5}$ concentration increased, indicating that both horizontal transport across Zhejiang's boundary and stable weather promoted the building up of $\text{PM}_{2.5}$ concentration.

High concentrations were transported from the region on the north of Zhejiang to Zhejiang by northerly wind before 7 December when the highest $\text{PM}_{2.5}$ concentration showed up, which was confirmed by the analysis of synoptic pattern. There were strong input fluxes mainly within the PBL and especially below altitude 500 m at the north

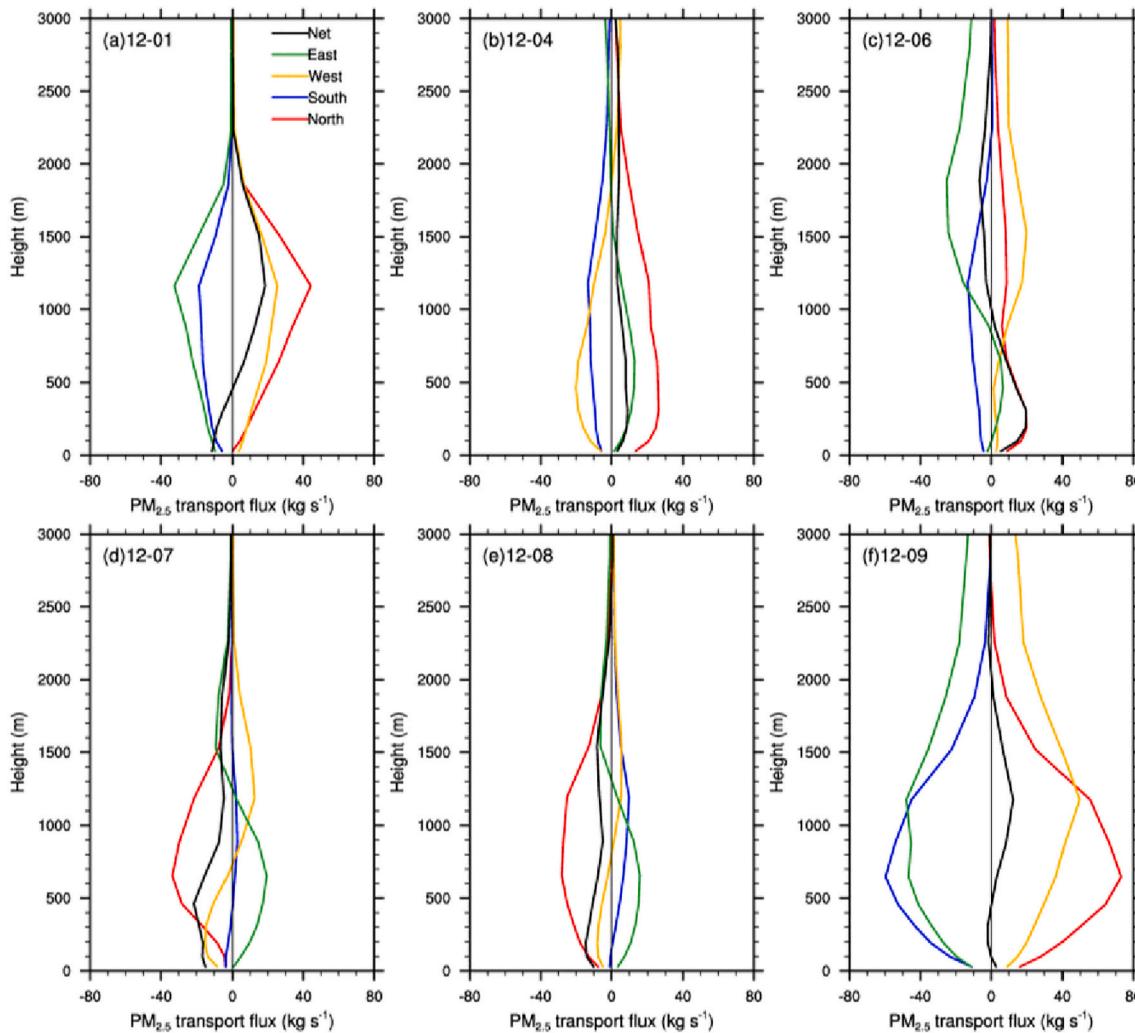


Fig. 11. Vertical profiles of the net transboundary transport flux of $\text{PM}_{2.5}$ over the north, south, west, east and the entire boundaries of Zhejiang during the selected days from 28 November to 10 December 2013.

boundaries on 4 and 6 December before the $\text{PM}_{2.5}$ peak concentration occurred on 7 December. After that, there were large output fluxes primarily above the PBL and especially between 400 m and 1200 m at the north and west boundaries transporting $\text{PM}_{2.5}$ out from Zhejiang. The analysis showed that the $\text{PM}_{2.5}$ transboundary transport across Zhejiang's boundary contributed 62% to the total $\text{PM}_{2.5}$ increase within Zhejiang during the period of severe haze formation from 3 to 6 December, and the primary emission and local secondary formation of

$\text{PM}_{2.5}$ contributed 15% and 23%, respectively. This indicates that the $\text{PM}_{2.5}$ transboundary transport from the surrounding areas to Zhejiang province was the main cause of such a severe haze event.

Declaration of Competing Interest

The authors declare no conflict of interest.

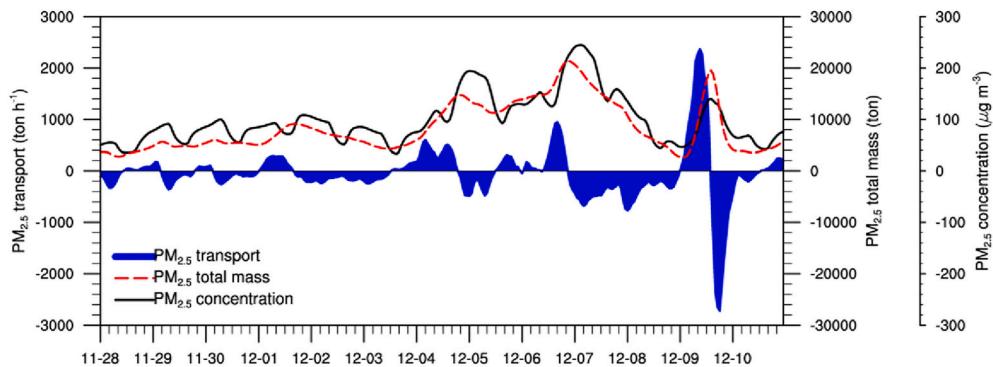


Fig. 12. Temporal variations of the transboundary transport flux of $\text{PM}_{2.5}$, total $\text{PM}_{2.5}$ suspended below 3 km height and the near-surface $\text{PM}_{2.5}$ concentration from 28 November to 10 December 2013.

Table 2

Components of conservation of mass formula for PM_{2.5} during different periods.
Units: ton.

	Case 1	Case 2
Components	11:00, 3 Dec-21:00 6 Dec	01:00-14:00, 9 Dec
ΔM	17,111.6	16,899.4
M_{Trans}	10,945.6	17,992.7
M_{Dep}	-473.6	-2349.4
M_{Emiss}	2697.8	427.7
M_{Chem}	3941.8	828.4
CR_T	62%	93%

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