Formation and Evolution Mechanisms of Severe Haze Pollution in the Sichuan Basin, Southwest China

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ABSTRACT

Severe haze episodes are important environmental issues, and the rapid formation and evolution mechanisms of such episodes over complex terrain remain poorly understood. The Sichuan Basin (SCB) periodically experienced heavy haze pollution during the winter of 2016, with the maximum regional average PM_{2.5} concentration reaching almost 120 $\mu\text{g m}^{-3}$. In this study, we characterize a severe haze episode in the SCB from 20 to 30 January 2017 using comprehensive measurements and model analyses. The evolution of this severe episode shows clear stages, with gradual PM_{2.5} increases under stagnant weather conditions in Stage I (aerosol accumulation stage) and with explosive PM_{2.5} increases mainly associated with cross-border transport from the southern SCB in Stage III (rapid formation stage). The process analysis results indicated that primary emissions and aerosol processes were the major sources of PM_{2.5} in these urban regions, whereas vertical transport and dry deposition generally acted as sinks of PM_{2.5}. In the presence of southwesterly synoptic winds, the aerosols emitted from the southern SCB were transported to Chengdu and the surrounding areas through horizontal transport and accounted for 66% of the PM_{2.5} concentration in Chengdu during Stage III. Our results reveal the detailed formation mechanism of a severe haze episode in the SCB under the effects of regional transport and synoptic forcing patterns to improve the understanding of haze formation in areas with complex terrain.

Keywords: Air quality; Haze pollution; Process analysis; Sichuan Basin.

INTRODUCTION

Due to rapid economic development and the acceleration of urbanization in China, regional haze pollution characterized by high fine particle (PM_{2.5}) concentrations has become a serious environmental issue (Chan and Yao, 2008; Lei *et al.*, 2011). To mitigate haze pollution, the “Air Pollution Prevention and Control Action Plan (APPCAP)” and the “National Ten Measures” were implemented by the Chinese State Council in 2013 to improve the air quality in China. Although the control measures have been generally successful,

large challenges remain, especially regarding the formation and evolution mechanisms of episodes of continuously high PM_{2.5}, which are poorly understood (Zheng *et al.*, 2015; Li *et al.*, 2017;).

Because of serious haze pollution resulting from high anthropogenic emissions and unfavorable meteorological conditions, the Sichuan Basin (SCB) has become one of the most polluted city clusters in China over the last few years (Zhao *et al.*, 2018). Extensive studies have been conducted to explore the source and formation of haze pollution in the SCB (Tian *et al.*, 2017; Qiao *et al.*, 2019a). Based on the National Centers for Environmental Prediction (NCEP) reanalysis data and clustering analysis method, Sun *et al.* (2020) classified the circulation patterns during periods of continuous pollution in the SCB and found that a weak high-pressure system located in the SCB could enhance pollution by suppressing the planetary boundary layer height (PBLH).

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By analyzing the chemical composition of $\text{PM}_{2.5}$, Song *et al.* (2019) reported that the secondary conversion of SO_2 , NO_x and nonmethane hydrocarbons (NMHCs) to sulfate, nitrate and secondary organic aerosols (SOAs), respectively, made significant contributions to persistent haze episodes in the SCB. The meteorological conditions in the SCB during winter are characterized by high relative humidity and low temperature, which favor the hygroscopic growth of fine particles and accelerate secondary transformations from gaseous precursors to inorganic ions (Kong *et al.*, 2020). Additionally, the complex basin topography of this region was found to increase the accumulation of $\text{PM}_{2.5}$ and intensify haze formation by reducing the wind speed and increasing the relative humidity (Zhang *et al.*, 2019). Several studies employed the CTM model to investigate the sources of $\text{PM}_{2.5}$. Zhang *et al.* (2016) utilized HJ-1 satellite data and simulation results from the WRF-Chem model to explore the formation of a haze event and found that low mean ventilation coefficients and decrease in PBLH were conducive to the occurrence of severe haze events. Using a source-oriented version of the Community Multiscale Air Quality (CMAQ) model, Qiao *et al.* (2019b) quantified the relative contributions from regions inside the SCB to the $\text{PM}_{2.5}$ concentration; they concluded that the transport of secondary inorganic aerosols (SIAs) and their precursors made significant contributions to haze pollution during the winter. However, previous studies focused on only monitored air pollutants, and the formation and evolution mechanisms of severe haze pollution in the SCB remain unclear. Consequently, thorough investigations involving the quantitative assessment of contributions from local emissions and regional transport to severe haze episodes are urgently needed (Chen *et al.*, 2014; Luo *et al.*, 2020).

In the present study, we integrate comprehensive monitoring data and the Weather Research and Forecasting-Community Multiscale Air Quality (WRF-CMAQ) modeling system to investigate the sources and formation mechanisms of an extreme regional haze episode in the SCB during the winter of 2016. The paper is organized as follows. In Sect. 2, the model configuration is described. In Sect. 3, the model performance is evaluated by comparison with observations. The formation and evolution mechanisms of the severe haze pollution are presented in Sect. 4. The conclusions are given in Sect. 5.

METHODS

Observational Data

The simulated meteorological parameters, including temperature, relative humidity, wind speed (WS) and wind direction, are compared with hourly observation data recorded at four typical national standard basic weather stations (Chengdu, Meishan, Ziyang and Neijiang), which were obtained from the China Meteorological Data Service Center (<http://data.cma.cn/en>). The hourly observed surface $\text{PM}_{2.5}$ concentrations over the SCB were collected from the China National Urban Air Quality Real-time Publishing Platform (<http://113.108.142.147:20035/>). The hourly mass concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , black carbon (BC), elemental carbon (EC) and organic carbon (OC) were

determined from samples collected on the roof of the Chengdu Environmental Science Research Institute (approximately 25 m above the ground) in Qingyang District, Chengdu ($30^\circ 56' \text{N}$, $104^\circ 05' \text{E}$), from 1 to 31 January 2017. This site (referenced as the CDAES site below) is a typical urban site that is close to the mixed commercial and residential area of Chengdu Second Ring Road (Fig. S1). At this sampling site, an in-situ gas and aerosol composition monitor (IGAC/S-611) was used to measure the water-soluble ions (SO_4^{2-} , NO_3^- , and NH_4^+) in $\text{PM}_{2.5}$ on an hourly basis. Previous studies have verified the accuracy of such monitoring through comparison with the commonly used annular denuder system, and the results showed good agreement (Young *et al.*, 2016; Tao *et al.*, 2018). Hourly concentrations of OC and EC in $\text{PM}_{2.5}$ were monitored using a Sunset Laboratory Model-4 OC-EC field analyzer with detection limits of $0.4 \mu\text{g C m}^{-3}$ for OC and $0.2 \mu\text{g C m}^{-3}$ for EC. The organic matter (OM) content was calculated by multiplying the OC content by a factor of 1.4. The hourly concentration of BC in $\text{PM}_{2.5}$ was monitored at a wavelength of 670 nm using a 5012 multiangle absorption photometer (MAAP, Thermo Fisher Corporation). Additional details were described in previous field studies (Song *et al.*, 2018; Yang *et al.*, 2020).

WRF-CMAQ Simulation

The Weather Research and Forecasting (WRF, version 3.9.1) model was applied to simulate meteorological fields, and CMAQ v5.2.1 was utilized to simulate air pollutants in the SCB during winter. The simulation period started on 15 January 2017 and ended on 31 January 2017. The first 5 days were discarded to account for spin-up and not included in the analysis. Fig. S2 illustrates the triple-nested domain with horizontal resolutions of 27 km, 9 km and 3 km. The outer domain covers all of East Asia, and the inner domain covers the SCB, as shown in Fig. 1. Here, we analyzed only the inner domain to minimize the bias from lateral boundary conditions. The vertical dimension had a resolution of 30 sigma layers.

The meteorological inputs for the WRF simulations were obtained from the NCEP Final (FNL) $1.0^\circ \times 1.0^\circ$ reanalysis data (<http://dss.ucar.edu/datasets/ds083.2/>). To improve the model performance, analysis nudging and observation nudging were adopted, and four-dimensional data assimilation (FDDA) data were obtained from the NCEP Automated Data Processing (ADP) Operational Global Surface (ds461.0) and Upper (ds351.0) Observations. The state variables for the water-vapor mixing ratio, potential temperature, and wind were nudged with strengths of 1×10^{-5} , 5×10^{-5} , and $5 \times 10^{-5} \text{ s}^{-1}$, respectively (Wu *et al.*, 2020; Xing *et al.*, 2017). The initial and boundary conditions for the CMAQ simulation were obtained from the MOZART-4 global chemical transport model (Emmons *et al.*, 2010). We obtained the anthropogenic emissions of air pollutants, with a grid resolution of $0.25^\circ \times 0.25^\circ$, from the Multiresolution Emission Inventory for China (MEIC) in 2016, which was developed by Tsinghua University (Zheng *et al.*, 2018). To reduce the uncertainty attributed to the outdated spatial surrogates in the spatial-temporal emission allocation process, we utilized the population distribution data from the LandScan 2017 database

Fig. 1. Average simulated surface $\text{PM}_{2.5}$ concentrations in domain I of China (a) and the Sichuan Basin (SCB) (b) in January 2017.

and GDP data from the database of the National Bureau of Statistics of China (NBSC) to allocate the emissions to the grid cells (Deng *et al.*, 2020). The Model of Emissions of Gases and Aerosols from Nature (MEGAN, version 2.1) was used to calculate the biogenic emissions (Guenther *et al.*, 2012). The options for the physical and chemical parameterizations of the WRF-CMAQ model are listed in Table 1.

Numerical Experiments

Four sensitivity experiments were performed to quantify the contributions of local emissions and regional transport during the studied episode, as shown in Table 2. In the BASE simulation, all anthropogenic and biogenic emissions were included. In the No-Anth experiment, only biogenic emissions were included. In the NoCD-Anth experiment, the anthropogenic emissions in Chengdu city were turned off. In contrast, we considered only anthropogenic emissions in Chengdu city in the OnlyCD-Anth experiment. Based on the above experiments, the relative contribution from regional transport was determined by comparing the simulation results of BASE to those of OnlyCD-Anth. The contribution from local emissions to the $\text{PM}_{2.5}$ concentration was identified by subtracting the NoCD-Anth results from the BASE results.

Process Analysis

Process analysis (PA) is a diagnostic method incorporated into the CMAQ model and includes integrated process rate (IPR) analysis and integrated reaction rate (IRR) analysis (Byun and Ching, 1999). The IPR method has been widely used to track the hourly relative contributions of physical/chemical processes to $\text{PM}_{2.5}$ concentrations in previous studies (Gao *et al.*, 2014; Fan *et al.*, 2015; Huang *et al.*, 2016). In this study, the IPR method was used to assess the processes that influence simulated $\text{PM}_{2.5}$ concentrations. The nine types of processes are gas-phase chemistry (denoted CHEM), emission

sources (denoted EMIS), aerosol processes (denoted AERO), cloud processes (denoted CLDS), dry deposition (denoted DDEP), horizontal advection (denoted HADV), horizontal diffusion (denoted HDIF), vertical advection (denoted ZADV) and vertical diffusion (denoted VDIF). It should be noted that horizontal transport (denoted HTRA) is defined as the sum of horizontal advection and horizontal diffusion, and vertical transport (denoted VTRA) is defined as the sum of vertical advection and vertical diffusion.

MODEL EVALUATION

Meteorological Parameters

We evaluated the performance of the WRF model to demonstrate the capability of the modeling system through verification statistics, including the normalized mean bias (NMB), the index of agreement (IOA), and the root mean square error (RMSE), for the hourly 2-m temperature (T2), 2-m relative humidity (RH2), 10-m wind speed (WS10) and 10-m wind direction (WD10) at the Chengdu, Meishan, Ziyang and Neijiang meteorological stations. As shown in Table 3, the simulated T2 and RH2 agreed well with the observations, with IOA values higher than 0.80, indicating that the model successfully captured the variation in T2 and RH2 in these four cities. For WS10 and WD10, the model displayed negative bias, with NMB values ranging from 16.69–42.36 and from 28.79–36.29, respectively. This bias may be attributed to the complex basin terrain and planetary boundary layer parameterization. Overall, these comparisons clearly demonstrate that the WRF model adequately reproduced the meteorological conditions.

$\text{PM}_{2.5}$ and its Chemical Components

Fig. 2 shows the time series of simulated hourly $\text{PM}_{2.5}$ concentrations in the BASE simulation and the observed $\text{PM}_{2.5}$ levels in the four cities from 20 to 30 January 2017.

Table 1. Modeling configuration used for the WRF-CMAQ simulations.

Model attribution	Configuration
Simulation period	15 January 2016 to 31 January 2017 (5-day spin-up period)
Domain	Sichuan Basin; Center = 30°N, 104°E
Horizontal resolution	27 km/9 km/3 km
Vertical resolution	30 layers from the surface to 50 hPa
Microphysics	Lin scheme
PBL physics scheme	Yonsei University (YSU) scheme
Shortwave and longwave radiation	Rapid Radiative Transfer Model for GCMs (RRTMG)
Land surface model	Noah land surface model
Gas-phase chemistry	Carbon bond chemical reaction mechanism (CB06)
Aerosol module/size	AERO6/3 modes

Table 2. Statistical metrics for comparing the observations and simulations.

Simulation	Description
BASE	All anthropogenic and biogenic emissions are included
No-Anth	Only biogenic emissions are included
NoCD-Anth	Anthropogenic emissions in Chengdu city are excluded
OnlyCD-Anth	Considers only anthropogenic emissions in Chengdu city

Table 3. Statistical metrics between the observations and simulations.

		Chengdu	Meishan	Zigong	Neijiang
2-m Temperature	NMB	−6.62	−3.69	−9.68	−11.36
	RMSE	4.16	2.12	6.47	6.81
	IOA	0.94	0.97	0.91	0.91
2-m Relative Humidity	NMB	−6.69	−10.49	−11.2	−13.2
	RMSE	15.81	14.64	12.28	16.49
	IOA	0.86	0.80	0.89	0.79
10-m Wind Speed	NMB	16.69	22.85	37.41	42.36
	RMSE	2.03	2.19	3.32	2.94
	IOA	0.72	0.62	0.66	0.69
10-m Wind Direction	NMB	28.79	36.29	35.78	29.46
	RMSE	46.97	56.81	42.23	49.44
	IOA	0.60	0.56	0.64	0.58

Generally, the CMAQ model reasonably reproduced the temporal evolution of the observed $\text{PM}_{2.5}$ concentrations in the four cities, with IOAs of 0.89, 0.84, 0.80, and 0.93 and mean biases of $-12.3 \mu\text{g m}^{-3}$, $-16.8 \mu\text{g m}^{-3}$, $-23.6 \mu\text{g m}^{-3}$ and $-5.8 \mu\text{g m}^{-3}$ for Chengdu, Meishan, Ziyang and Neijiang, respectively. In general, the CMAQ model captured the peaks and temporal variations in the $\text{PM}_{2.5}$ concentrations from 20 to 28 January in these cities but underestimated the peak $\text{PM}_{2.5}$ concentration on 28 January in Zigong.

Fig. 3 compares the simulated and observed OM , SO_4^{2-} , NO_3^- , and NH_4^+ concentrations at the CDAES site averaged from 20 to 30 January 2017. The CMAQ model underestimated the concentrations of OM , SO_4^{2-} and NO_3^- by $0.8 \mu\text{g m}^{-3}$, $2.3 \mu\text{g m}^{-3}$, and $5.1 \mu\text{g m}^{-3}$, respectively, but overestimated the NH_4^+ concentration by $1.3 \mu\text{g m}^{-3}$. For OM , the underestimation may have been attributed to the lack of SOA parameterizations in the AERO06 aerosol module. The bias associated with SIAs (including NH_4^+ , SO_4^{2-} and NO_3^-) has also been noted in previous studies and potentially derives from the lack of heterogeneous reactions in the chemistry mechanisms in the models, the allocation bias for emission

inventories, and the failure to capture relevant changes in meteorological conditions (Zheng *et al.*, 2015).

RESULTS AND DISCUSSION

Spatiotemporal Evolution of Severe Haze Formation

The weather conditions and air pollutant concentrations in four highly polluted cities during this episode are presented in Fig. 4. During this severe haze episode, the $\text{PM}_{2.5}$ concentrations in these cities all reached peak values on January 28th; notably, the surface $\text{PM}_{2.5}$ concentration rapidly increased from $100 \mu\text{g m}^{-3}$ to more than $600 \mu\text{g m}^{-3}$ in less than a day in Neijiang and Zigong; these peaks were the highest values recorded in 2017. The weather conditions in the SCB were characterized by persistent high relative humidity greater than 90% and low WS10 (less than 3 m s^{-1}) during this period.

Fig. 5 shows the spatial distributions of the simulated daily mean surface $\text{PM}_{2.5}$ concentrations within the SCB from 20 to 28 January 2017. Before the haze episode (e.g., on January 20), the daily $\text{PM}_{2.5}$ concentration in most areas

Fig. 2. Comparison of the observed (blue line) and simulated (black dots) hourly surface $PM_{2.5}$ concentrations in four cities in the Sichuan Basin (SCB) from 20 January to 30 January 2017.

Fig. 3. Comparison of the observed and simulated surface mass concentrations ($\mu g m^{-3}$) of SO_4^{2-} (red), NO_3^- (blue), NH_4^+ (purple), OM (orange) at CDAES site (30°56'N, 104°05'E) averaged over 20–30 January 2017. Mean biases (MB, unit: $\mu g m^{-3}$) are also listed for each species (colored numbers).

of the SCB was less than $100 \mu g m^{-3}$ under a prevailing clean southerly air flow. On 21 January, a uniform pressure field was present over the basin, and the regional average WS was less than $3 m s^{-1}$. The boundary layer became stable (see Fig. S3), which was favorable for constraining the aerosol

particles within the low mixing layer. Consequently, a high $PM_{2.5}$ belt formed over the western SCB, with the daily mean $PM_{2.5}$ concentration exceeding $140 \mu g m^{-3}$. During this stage, a uniform pressure field continuously controlled most regions of the SCB, resulting in $PM_{2.5}$ concentrations higher than $120 \mu g m^{-3}$. However, on 26 January, the SCB was affected by effective convection caused by synoptic conditions, resulting in a temporary decrease in the concentration of $PM_{2.5}$ due to wet scavenging by precipitation. On 27 January, the weather pattern over the SCB was characterized by strong anticyclonic circulation. Moreover, the SCB is located behind a ridge centered in eastern Tibetan (Fig. S4(g)), which causes the prevailing regional wind field to transition to strong southeasterly winds. Therefore, air masses were transported from south to north, leading to a significant increase in the $PM_{2.5}$ concentration in Chengdu and the surrounding areas, with the maximum daily mean value exceeding $270 \mu g m^{-3}$. After 28 January, the SCB was affected by northerly winds that brought dry and clean air masses into the basin. The $PM_{2.5}$ concentrations in most areas of the SCB decreased to 40 – $100 \mu g m^{-3}$.

Based on the evolution pattern of simulated $PM_{2.5}$ concentrations over the SCB, we divide this episode into four stages:

- (I) The aerosol accumulation stage (20–25 January). In Stage I, the daily mean $PM_{2.5}$ concentrations averaged over the SCB slowly increased from 30.8 to $240.7 \mu g m^{-3}$, and these levels persisted for five days.
- (II) The clearance stage (26 January). The $PM_{2.5}$ concentrations in the SCB decreased to $80 \mu g m^{-3}$, which was caused by precipitation.
- (III) The rapid formation stage (27–28 January). During this

Fig. 4. Evolution of the observed $PM_{2.5}$, PM_{10} , and gaseous pollutant concentrations; 2-m temperature (T2); 2-m relative humidity (RH2); 10-m wind speed (WS10); and 10-m wind direction (WD10) in January 2017 in four cities: (a) Chengdu, (b) Meishan, (c) Neijiang, and (d) Zigong.

stage, the daily average $PM_{2.5}$ concentrations in most cities of the SCB reached the “heavily polluted” air quality threshold value (24-h average $PM_{2.5}$ concentration $> 150 \mu g m^{-3}$). In Neijiang and Leshan, the $PM_{2.5}$ concentration even reached 897 and 724 $\mu g m^{-3}$, respectively.

(IV) The aerosol dispersion stage (29–31 January). In this stage, the $PM_{2.5}$ concentration gradually decreased dramatically to low levels (50–72 $\mu g m^{-3}$) with increasing WS10.

Contributions from Local Emissions versus Regional Transport

Previous studies have indicated that local anthropogenic emissions are the major source of high $PM_{2.5}$ concentrations during severe haze episodes in the SCB (Fan *et al.*, 2020). Despite the dominant contribution from local emissions, the contribution of regional transport to $PM_{2.5}$ concentrations has not been fully quantified in the SCB (Zhao *et al.*, 2019). Regional transport is responsible for more than 30% of the $PM_{2.5}$ concentration variations in the SCB during winter

Fig. 5. The spatial distributions of simulated daily $\text{PM}_{2.5}$ concentrations (shaded, $\mu\text{g m}^{-3}$) and wind vectors (arrows, m s^{-1}) from 20 to 28 January 2017

(Qiao *et al.*, 2019b). In this section, we discuss the contributions of local anthropogenic emissions and regional transport to the $\text{PM}_{2.5}$ concentration in the SCB to determine the relative importance of each factor during the studied severe haze episode.

Fig. 6 illustrates the relative contributions of local emissions and regional transport to the simulated $\text{PM}_{2.5}$ level in Chengdu city. The episode featured different evolution processes, with a gradual $\text{PM}_{2.5}$ increase in Stage I and an explosive $\text{PM}_{2.5}$ increase in Stage III. In Stage I, the stagnant conditions near the surface and low WS10 were not conducive to the dispersion of air pollutants, which led to the local accumulation and chemical production of secondary pollutants. Therefore, the relatively high $\text{PM}_{2.5}$ concentration

($141.1 \mu\text{g m}^{-3}$) was principally caused by local emissions during Stage I. In Stage II, the $\text{PM}_{2.5}$ concentration in Chengdu was mainly associated with the combined effects of local emissions and regional transport. The contributions of local emissions and regional transport to the $\text{PM}_{2.5}$ concentration were comparable in this stage (44% versus 52%, respectively). The cross-border transport of polluted air masses from the southern SCB under prevailing southeasterly winds was the main contributor to haze formation in Stage III, with the contribution of regional transport reaching 66% and that of local emissions reaching 32%. During Stage IV, the $\text{PM}_{2.5}$ levels in Chengdu were dominated by local contributions (58%), and the contribution from regional transport decreased to 38%.

Fig. 6. Contributions of local emissions (red) and regional transport (blue) to the surface PM_{2.5} concentrations averaged over Chengdu from 20 to 30 January 2017. The absolute contributions (μg m⁻³) are shown using bars, and the percent contributions (%) are shown using lines. The PM_{2.5} concentration and the percent contributions averaged over each stage are listed at the bottom of the figure. Note: The “others” contribution represents the PM_{2.5} concentration biased from the brute force method.

However, it should be noted that the brute force method used in this study to attribute the contribution of local emissions and regional transport has limitations due to nonlinearities of the chemistry of PM_{2.5} formation (Clappier *et al.*, 2017; Thunis *et al.*, 2019). Although the brute force method has been widely used in estimating the contributions of PM_{2.5} sources (Kim *et al.*, 2017; Chen *et al.*, 2019; Cheng *et al.*, 2019), further investigations using source-oriented methods such as CAMx-PSAT and CMAQ-ISAM should be considered to reduce the nonlinear effects in future studies.

Process Analysis of Variations in the PM_{2.5} Concentration

Fig. 7 shows the hourly contributions of individual atmospheric physical and chemical processes to variations in the surface PM_{2.5} concentration over Chengdu in different stages. In the beginning of Stage I, EMIS was the dominant source of surface PM_{2.5} in Chengdu and was responsible for 72.8% of the accumulated PM_{2.5}. Compared with the dominant effect of EMIS on the PM_{2.5} concentration, the contribution of AERO to surface PM_{2.5} was relatively small, accounting for only ~8.8% of the accumulated PM_{2.5} mass concentration. The contribution of CLDS was almost negligible in the surface layer. DDEP, HTRA and VTRA were the major removal pathways of PM_{2.5}, accounting for ~6.6, ~14.8% and ~76.4% of the removal, respectively. The maximum positive contribution of EMIS was observed during rush-hour periods (07:00–08:00 and 16:00–19:00), whereas the maximum negative contributions of VTRA appeared at midnight. The PA results in the middle of Stage I (January 21–22) differed from those at the beginning of Stage I. Even though EMIS still acted as the predominate contributor to PM_{2.5}, HTRA gradually became the second largest source of surface PM_{2.5}, accounting for 24.3%. In Stage II, the contribution of AERO to the surface PM_{2.5} was quite low

compared with that in Stage I. Notably, the PM_{2.5} concentration decreased dramatically during this stage, which can be attributed to wet deposition as a result of precipitation events (as discussed before). When the episode reached Stage III, the contribution from HTRA increased dramatically, as an important source of PM_{2.5} in addition to primary emissions. This phenomenon reflected the important role of regional PM_{2.5} transport from the southern SCB to Chengdu and the surrounding areas under the prevailing wind fields (see Fig. 5(h)–5(i)). Additionally, VTRA still had a negative influence on the PM_{2.5} concentration and was responsible for ~88% of the reduction. It should be noted that the maximum negative contributions of VTRA in Stage III appeared at night due to the strong southeasterly winds. During Stage IV, although EMIS still acted as a stable source of PM_{2.5}, HTRA became the main process of PM_{2.5} removal, leading to a gradual decrease in the PM_{2.5} concentration. In summary, the rapid increase in PM_{2.5} in Chengdu was mainly attributed to HTRA, which reflected the influence of regional transport from the southern and southeastern SCB. Furthermore, the essential role of primary EMIS, which acted as a stable positive contributor to PM_{2.5}, cannot be neglected.

CONCLUSIONS

In this study, we comprehensively investigate the formation and evolution mechanisms of a severe regional haze episode that occurred over the SCB from 20 January 2017 to 31 January 2017. The roles of local emissions and regional transport in the haze episode were identified by sensitivity experiments. Additionally, the IPR method was used to quantify the contributions of each physical/chemical process to the variation in the PM_{2.5} concentration.

Four haze stages were identified, with a record-breaking hazy episode with a peak concentration of nearly 400 μg m⁻³

Fig. 7. Hourly contributions of individual processes to the $\text{PM}_{2.5}$ mass concentration in Chengdu from 20 January 2017 to 31 January 2017.

was observed on 27 January. Sensitivity experiments indicated that the local emissions in Chengdu accounted for 46% of the total $\text{PM}_{2.5}$ concentrations on average, thus acting as a stable positive contributor. In Stage I, the concentration of surface $\text{PM}_{2.5}$ was controlled by local emissions (68%). The contribution of regional transport from the areas within the simulation domain, excluding Chengdu, increased significantly (Stages II and III), which was the main cause of the persistent severe episode. Therefore, joint emission control is the most effective way to mitigate regional haze pollution.

Based on the IPR analysis, emissions (EMIS) and aerosol chemistry (AERO) were the major sources of pollutants and were responsible for 1.3% and 5.2% of the $\text{PM}_{2.5}$, respectively. In contrast, the main sinks were VTRA and DDEP during the study period, accounting for 9.4% and 5.2% of reduction, respectively. However, the substantial $\text{PM}_{2.5}$ increase in Chengdu during the rapid formation stage was attributed to the positive contribution of HTRA, indicating that regional transport is significant in regional haze formation. This work and previous studies all highlighted the essential role of cross-border transport between cities inside SCB in wintertime haze episodes. To further promote our understanding on severe haze episodes occurred over SCB in wintertime, future work should be considered on elucidating the formation of organic aerosols under high RH and cloudy conditions. Additional work is also needed to identify the contribution from different source regions to the $\text{PM}_{2.5}$ and major transport pathways at different heights in order to further understand the regional transport mechanism (Huang *et al.*, 2020).

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <https://doi.org/10.4209/aaqr.2020.04.0173>

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