Impact of Atmospheric Circulation and Meteorological Parameters on Wintertime Atmospheric Extinction in Chengdu and Chongqing of Southwest China during 2001–2016

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ABSTRACT

This study investigated the effect of atmospheric circulation and meteorological parameters on the surface atmospheric extinction coefficient (SEC) in Chengdu and Chongqing of southwest China during the winters of 2001–2016. Four predominant circulation types (CT) were first identified, representing a high-pressure system in the north (CT 1), a low-pressure system in the north (CT 2), a weak high-pressure system (CT 3) and a local low-pressure system (CT 4) at a geopotential height of 850 hPa. In Chengdu, variation in the SEC between the CTs was dominated by meteorological factors, such as wind, subsidence, inversion and planetary boundary layer height (PBLH), whereas the variation within each CT was mainly controlled by dispersion-related parameters (such as wind speed and PBLH for CTs 1, 2 and 4, which were associated with strong weather systems) or chemistry-related parameters (such as RH and T for CT 3, which was associated with a weak system). In Chongqing, dispersion and chemical processes were equally important due to the weaker impact of the atmospheric circulation. A new method for discriminating between synoptic and non-synoptic signals in SEC time series was proposed to estimate the effect of the circulation intensity by considering multiple influential meteorological parameters. Atmospheric circulation heightened the interannual SEC variations by ~23% in both Chengdu and Chongqing in more than half of the years but reduced them by 44.79% and 8.02% in Chengdu and Chongqing, respectively, in the remaining years. Synoptic changes were estimated to contribute 5–10% of the decreasing SEC trend but less than 1% of the increasing trend.

Keywords: Atmospheric extinction coefficient; Atmospheric circulation; Meteorological parameters.

INTRODUCTION

The deterioration of air quality in China, mainly caused by high aerosol loading associated with the rapidly increasing energy consumption, industrialization and urbanization, has been a severe environmental issue in the recent decades (Chan and Yao, 2008). Surface atmospheric extinction coefficient (SEC) is widely used as a proxy of particulate pollution (Li *et al.*, 2016). SEC can be inferred from horizontal visibility (VIS) (Che *et al.*, 2007), scattering and absorption coefficient (He *et al.*, 2009), or column integrated aerosol optical depth (AOD) (Lin *et al.*, 2015).

The level of particulate pollution is controlled by emission intensity, secondary aerosol formation, dispersion, transport and removal of pollutants. All these processes are strongly affected by meteorological parameters, including wind speed (WS), wind direction (WD), relative humidity (RH), temperature (T) and planetary boundary layer height (PBLH). Weak wind is unfavorable for the dispersion, transport and dry deposition processes (Zhang *et al.*, 2001; Zhang *et al.*, 2015a; Wang *et al.*, 2016), hence responsible for the accumulation of air pollutants. WD controls the direction of horizontal transport and makes different air quality conditions associated with surrounding regions (He *et al.*, 2009; Zhang *et al.*, 2015a). RH and T are associated with the gas-to-particle conversion process by shifting the partition of semi-volatile species into the aerosol phase (Hu *et al.*, 2008; Zhang *et al.*, 2015b). Low PBLH, generally accompanied with atmospheric inversion and subsidence,

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suppresses vertical mixing and causes high concentration of pollutants (Wu *et al.*, 2013; Petaja *et al.*, 2016). The meteorological parameters affecting air quality are interrelated and modulated by synoptic-scale circulation (Zhang *et al.*, 2012). Under the condition of quasi-stable emission, circulation pattern will be the predominant factor causing day-to-day variations of air pollutants. The impact of atmospheric circulations on air quality has been investigated for pollution episodes in various areas of China. For example, stable and weak anti-cyclone synoptic condition was in favor of heavy air pollution formation (Wei *et al.*, 2011; Liu *et al.*, 2013) and variant circulation patterns controlled the accumulation, spikes and dissipation of pollutants (Chen *et al.*, 2008) in northern China. Similarly, in eastern China pollution episodes were associated with the uniform pressure field and anti-cyclone circulation, while clean episodes occurred when strong southeastward cold air advection prevailed in lower troposphere (Zheng *et al.*, 2015). In southern China, poor air quality was typically associated with the warm period before a cold front and the subsidence period controlled by a tropical cyclone (Feng *et al.*, 2007; Wu *et al.*, 2013; Yan *et al.*, 2016). However, the impact of atmospheric circulation on air quality in southwestern China has rarely been investigated.

In addition to the case studies mentioned above, circulation type classification (CTC) is another well-established tool for analyzing the relationships between the circulation patterns and environmental variables (Philipp *et al.*, 2010). Through implementing CTC, the time series of circulation fields, such as pressure and wind, are categorized as several circulation types (CT) based on the similarity between these fields. Accordingly, the time series of environmental variables are grouped into different CTs. One advantage of doing CTC is that certain CT represents typical weather condition (combination of meteorological parameters). Hence, it is quite straightforward to investigate the influence of circulation on air quality by analyzing the difference of environmental variables between different CTs. Zhang *et al.* (2012) identified nine CTs and analyzed the air quality and pollutant transport pathways for each CT in Beijing in northern China. Zhang *et al.* (2013) classified seven CTs and found that the cyclone-related CTs were associated with relatively higher O₃ concentrations and variations in Hong Kong. Ye *et al.* (2016) used CTC to examine the impact of circulation patterns on planetary boundary structures and air quality in North China Plain.

The interannual variations and long-term trends of air quality are determined by the variations in both air pollutant emission and atmospheric circulation. One advantage of doing CTC is that it provides a technique to discriminate the circulation related signals from other signals. Comrie (1992) presented a method to decompose environmental data into its synoptic and non-synoptic components using occurrence frequencies of CTs. Hegarty *et al.* (2007) further considered both the intensity and frequency of each CT and demonstrated that synoptic signal contributed 46% of the interannual variability in summertime O₃ in the northeastern United States. Zhang *et al.* (2012) found that 54% of the visibility improvements were attributed to the

CT frequency anomaly during the 2008 Olympics in Beijing. Zhang *et al.* (2013) presented that 36% of the observed decadal increasing trends of O₃ in Hong Kong were results of the interannual variability in frequencies and intensities of circulation patterns.

Theoretically, it is reasonable to consider synchronously intensities and frequencies of CTs while discriminating between interannual synoptic and non-synoptic signals. (Hegarty *et al.*, 2007). For each CT, the anomalies of environmental variables (ΔX , defined as the difference between the environmental variables for a given year and multi-year average within the study period) were linearly correlated to certain predefined circulation intensity index (CII). Hence, for each year ΔX could be derived from CII and added to the synoptic signals from CT frequency only (Hegarty *et al.*, 2007). However, several issues still need to be addressed in these procedures. Firstly, ΔX contains information about non-synoptic factors. Specifically, in China the dramatic interannual variations of anthropogenic emissions should contribute to ΔX significantly. Secondly, single CII seems not to be fully representative of the intensity of CT. For instance, Zhang *et al.* (2013) provided four possible indexes, including the domain-averaged sea level pressure, the central pressure of a dominant system, the pressure gradient, and the effective intensity, and eventually selected the pressure gradient and effective intensity as CII for high- and low-pressure system, respectively, based on the best correlation with O₃ anomalies. Apparently, the contribution of indexes with weaker correlation was ignored, suggesting the need of improving this method.

Chengdu and Chongqing are the two biggest cities in Sichuan Basin (southwestern China) and acknowledged as one of the most polluted regions in China (Wu *et al.*, 2012; Wang *et al.*, 2014; Meng *et al.*, 2015; Zhao *et al.*, 2016b; Tao *et al.*, 2017). As shown in Fig. 1, the altitude is lower than 500 m in the basin areas and exceeds 1000 m at the basin edges. The annual average visibilities of these cities have been less than 10 km since the 1970s (Chen and Xie, 2012) and aerosol pollution events occurred mainly in winter (Wang *et al.*, 2014; Che *et al.*, 2015; Zhao *et al.*, 2016b; Wang *et al.*, 2017). Since the West Development Project launched in 2000 by the Chinese government, energy consumption and corresponding pollutant emission have increased dramatically. Meanwhile, stringent air pollution control measures have been taken, such as removal of “yellow label” vehicles and migration of industries away from urban areas (Wang *et al.*, 2017). These factors, accompanied with the effects of climate change and urbanization (Miao *et al.*, 2015; Zhao *et al.*, 2016a), complicated recent air quality trends in Chengdu and Chongqing. For example, satellite-observed AOD reached the maximum value in 2010 and then started decreasing in the past decade in this region (Liu *et al.*, 2016). Similarly, atmospheric extinction shifted from increasing trend to decreasing one around 2006 in southwestern China (Li *et al.*, 2016). The goals of this study are to (1) identify the main atmospheric circulation patterns influencing Sichuan Basin in winter; (2) investigate the relationships between

Fig. 1. Terrain height of Sichuan Basin and the location of Chengdu and Chongqing.

atmospheric circulation patterns, wintertime atmospheric extinction and meteorological parameters in Chengdu and Chongqing; (3) develop an improved method to discriminate synoptic signals from non-synoptic signals in time series of atmospheric extinction; and (4) examine the contribution of synoptic signals to the variations of wintertime atmospheric extinction in these two cities during 2001–2016.

DATA AND METHODS

Meteorology and Visibility Data

The study area contains mountain regions with high elevations. The 850 hPa geopotential height (HGT) data, instead of commonly used mean sea level pressure, were thus used to drive the CTC. The gridded 850 hPa HGT fields for the period of December 2000 to February 2016 were extracted from the National Center for Environmental Prediction (NCEP) FNL (Final) operational global analysis data (in $1^\circ \times 1^\circ$ horizontal resolution). All the HGT fields covering the domain of $20^\circ\text{--}40^\circ\text{N}$ and $90^\circ\text{--}120^\circ\text{E}$ at 00 UTC in December, January and February were used in the CTC analysis. Besides, the PBLH fields from the FNL data at 06 UTC were used to represent boundary layer development during daytime while the three-dimensional vertical wind speed fields at 00 UTC were also obtained to analyze atmospheric subsidence.

The hourly visibility and meteorology data of the Integrated Surface Database provided by NOAA's National Centers for Environmental Information (available at <https://www1.ncdc.noaa.gov/pub/data/noaa/>) for 2000–2016 were used in this study. Station 562940 and 575150 (shown as triangles in Fig. 1) were selected to represent Chengdu and Chongqing, respectively. Following the

quality control strategy described in Li *et al.* (2016), only the surface synoptic observations (SYNOP) at 3 h intervals were used. The extracted variables included the visibility, temperature, dew point (T_d), wind speed and direction, and precipitation. Based on these variables, RH (in %) was calculated from T (in $^\circ\text{C}$) and T_d (in $^\circ\text{C}$) using

$$\text{RH} \approx \exp \left[17.27 \left(\frac{T_d}{T_d + 237.30} - \frac{T}{T + 237.30} \right) \right] \quad (\text{Murray, 1967}).$$

The hygroscopic growth of aerosols was corrected using the calculated RH according to the algorithm described in Che *et al.* (2007). The effect of RH on extinction coefficient depends on the chemical composition of aerosol. The composition of wintertime aerosol between 2002–2014 in Sichuan Basin were seldom reported. By implementing a revised IMPROVE algorithm (Pitchford *et al.*, 2007; Wang *et al.*, 2017) using observed aerosol composition in 2002 (Wang *et al.*, 2004; Yang *et al.*, 2011) and 2014–2015 (Wang *et al.*, 2018), we evaluated the upper limits of the relative errors caused by hygroscopic growth correction. The upper limits of relative errors ranged from 12.49% (CT 4) to 24.75% (CT 3) in Chengdu and around 11% in Chongqing (see section S1 of the supplementary materials for the details). Then the SEC (in km^{-1}) was calculated from the corrected VIS (in km) using the formula $\text{SEC} = 3.912/\text{VIS}$ (Koschmieder, 1926; Li *et al.*, 2016). Note that the SEC represent the atmospheric extinction coefficient including gaseous and aerosol contributions. Considering that the contribution of gases was likely less than 5% in Chengdu and Chongqing (Wang *et al.*, 2018), the variations of atmospheric extinction were analyzed without considering gaseous contributions in this research. Although it is mostly dry in winter in Chengdu

and Chongqing, the potential influence of precipitation needs to be eliminated. Any data associated with RH higher than 90% were first excluded. The whole-day data with daily precipitation greater than 5 mm were then excluded. Daily, seasonal and within-CT averages were only calculated if more than 50% of the data were kept after applying the above criteria.

The temperature profiles at 00 UTC were obtained from the Integrated Global Radiosonde Archive (IGRA) of NOAA to explore atmospheric inversion. The IGRA records radiosonde observations at globally distributed stations. The available observations in Chongqing were at Station 575150 for the entire study period, while in Chengdu they were at Station 562940 for the time period before 2005 and at Station 562187 afterwards. The distance between the two stations in Chengdu is about 20 km. The low level atmospheric inversion directly affects the vertical dispersion of air pollutants. Hence, we focused on the inversion occurred below 1500 m above ground level. The vertical resolution of IGRA profiles, with 3–5 layers between 0 and 1500 m, was too low to obtain the detailed structure of temperature profiles. We thus defined the lowest inversion bottom as the height of the lowest layer at which the temperature was lower than that at the upper layer. The temperature gradient at the lowest inversion bottom was calculated using the temperatures at these two adjacent layers. The occurrence frequency, vertical distribution of the lowest inversion bottom and temperature gradient were examined for each CT.

Circulation Type Classification

Several algorithms exist in literature for classifying circulation types in climatology studies. A comparison of these algorithms did not find any superiority of one algorithm over another statically (Philipp *et al.*, 2016). Based on the successful application of T-mode Principal Component Analysis (PCA) method in previous studies, we chose the PTT (T-mode PCA orthogonally rotated) method integrated in the software package cost733class (available at <http://cost733.geo.uni-augsburg.de>) to do CTC in this study. The 850 hPa HGT of grids with altitudes above 3000 m were excluded in CTC processes to diminish the potential vertical extrapolation errors.

Correlation Strategy for Multi-year Observations

The Pearson correlation was implemented to examine the relationships between daily averaged SEC and local meteorological parameters for each CT. However, for multi-year observations the correlations might be overwhelmed by the interannual variations of SEC and meteorological parameters. The original daily averaged time series have to be detrended before correlation analysis (Tai *et al.*, 2010; Li *et al.*, 2014). In this study, the relative anomalies (V) of SEC and meteorological parameters were invoked in the correlation and defined as:

$$V_{X,dnm} = \frac{X_{dnm} - \overline{X_{nm}}}{\overline{X_{nm}}}, \quad X = \text{SEC, RH, WS, PBLH, or T} \quad (1)$$

where $V_{X,dnm}$ is the variable at the d-th day in year n (e.g., year 2001 means from December 2000 to February 2001) for the m-th CT, and $\overline{X_{nm}}$ is seasonal mean of X in year n for the m-th CT.

Modified Method for Discriminating Synoptic Signals from Non-synoptic Signals

The seasonal mean SEC time series for the winters from 2001 to 2016 (observed signal, $\overline{\text{SEC}_n}$) were decomposed into overall synoptic signals (S_n) and non-synoptic signals (E_n). Following Comrie (1992) and Hegarty *et al.* (2007), the synoptic signals have the following form:

$$S_n = \frac{\sum_{m=1}^M (\overline{\text{SEC}_m} + \Delta \text{SEC}_{nm}) F_{nm}}{\sum_{m=1}^M F_{nm}} \quad (2)$$

where F_{nm} is the number of occurrence days in year n for the m-th CT, $\overline{\text{SEC}_m}$ is the 16-year winter average of SEC for the m-th CT, and ΔSEC_{nm} is the anomaly of seasonal mean SEC in year n for the m-th CT. On the right side of Eq. (2) the first term corresponds to the interannual variations of the occurrence frequencies (synoptic occurrence signal, O_n), and the second term to the within-type variations of circulation intensity and meteorological parameters.

In Hegarty *et al.* (2007) and Zhang *et al.* (2013), ΔSEC_{nm} was calculated from a linear fit between SEC anomalies and selected CII for each CT. Here, two modifications were made on the ΔSEC_{nm} term. One was that the linear fitting was implemented for detrended time series (relative anomalies) of SEC and then ΔSEC_{nm} was calculated from the derived relative anomalies. The other one was that a number of meteorological parameters, instead of single CII, was used to represent the within-CT variations of circulation intensity. The main steps of calculating ΔSEC_{nm} term are listed in section S2 of the supplementary materials.

According to the results of Comrie (1992), the decreases in the synoptic signals equal to the increases in the non-synoptic signals. Hence, the derived non-synoptic signals can be determined by:

$$E_{2001} = \overline{\text{SEC}_m} - S_{2001} \quad (3)$$

$$E_n = E_{n-1} + (\overline{\text{SEC}_n} - \overline{\text{SEC}_{n-1}}) - (S_n - S_{n-1}), \quad n = 2002, \dots, 2016 \quad (4)$$

RESULTS AND DISCUSSION

Main Circulation Patterns Influencing the Sichuan Basin

A total of eight CTs were obtained by adopting the CTC processes for the winters during 2001–2016 over the Sichuan Basin, but 98.8% of all the days were belonged to four of the eight CTs. Hence, the four dominant circulation patterns (noted as CT 1–4 below) were selected for further analysis. The characteristics of 850 hPa HGT, sea-level pressure (SLP) and 10 m wind fields are depicted in Fig. 2. CT 1 occurred most frequently, accounting for 38.6% of the winter days, while CTs 2–4 accounted for 21.1%,

Fig. 2. Circulation characteristics of the predominant CTs for the winter in 2001–2016. The colored shadings and black contour lines are the within-CT mean 850 hPa geopotential height in gpm and mean sea-level pressure in hPa, respectively. The gray vectors are the 10-m wind fields. The triangles are the locations of Chengdu (west) and Chongqing (east). The number in the title is the occurrence frequencies of corresponding CT. The dashed square represents the region of Sichuan Basin. The blank area is the plateau region with altitude higher than 3000 m.

19.7% and 19.5%, respectively. For CT 1, a strong high pressure was centered to the north of Sichuan Basin in terms of both 850 hPa HGT and SLP. For CT 2, Sichuan Basin is located at the southeast edge of a low-pressure system at 850 hPa and in the rear of a high-pressure system at sea level. CT 3 was associated with weak and unique high-pressure system prevailing from sea level to 850 hPa. CT 4 represented local low-pressure system controlling at 850 hPa and high-pressure system to the north at sea level. Due to the basin topography, the 10 m wind velocity in Sichuan Basin was distinctly lower than the surrounding areas (Ye and Gao, 1979). Within the basin, the wind velocity in the plain regions was higher than that in the mountainous regions. The average patterns of wind direction in Sichuan Basin were similar for all CTs, showing cyclonic circulations.

Impact of Circulation Patterns on SEC and Meteorological Parameters

The means, medians, 25th and 75th percentiles, and extreme limits of SEC for each CT in Chengdu and Chongqing are presented in Fig. 3 while the corresponding mean values of SEC, RH, WS, PBLH and T are listed in Table 1. In Chengdu, CT 3 had the highest and CT 4 had the lowest

SEC with a mean value (with $\pm 1 \sigma$) of 1.09 ± 0.67 and $0.73 \pm 0.51 \text{ km}^{-1}$, respectively. The mean SEC for CT 2 ($1.02 \pm 0.85 \text{ km}^{-1}$) was slightly lower than that for CT 3. The CT 1 was related to medium pollution levels, with the mean SEC of $0.84 \pm 0.73 \text{ km}^{-1}$. In Chongqing, the same descending order of mean SEC was observed, i.e., CT 3 (1.10 ± 0.80) > CT 2 (1.02 ± 0.57) > CT 1 (1.00 ± 0.55) > CT 4 (0.89 ± 0.54), but with smaller between-CT differences than those in Chengdu. These averages for different CTs were significantly different at 0.05 level except for CT 2 and CT 3 in Chengdu and CT 1 and CT 2 in Chongqing. The percentiles showed similar patterns in terms of between-CT differences in most cases. The only exception was between CT 2 and CT 3 in Chengdu which showed opposite trends between the median and mean values, i.e., CT 3 had lower median but higher mean than CT 2, implying more heavily polluted events from CT 3 than CT 2 in Chengdu. Furthermore, the probability distribution of SEC showed higher SECs for CT 3 and CT 2 (see section S3 of the supplementary materials). In conclusion, the weak high-pressure system (CT 3) and local low-pressure system (CT 4) were responsible for the highest and lowest SEC, respectively. When the study areas were on the edge of northerly centered high (CT 1)- or low (CT 2)-pressure

Fig. 3. The surface extinction coefficient for each CT in Chengdu and Chongqing. The upper limit, center bar, lower limits of the box are 25th, 50th and 75th percentiles, respectively. The upper and lower whiskers are the maximum and minimum values. The dots represent the average values.

systems, air pollution was in medium level compared to that for CT 3 and CT 4.

The frequency and mean wind speed (WS) from 16 wind directions for each CT in Chengdu and Chongqing are presented in Fig. 4. In Chongqing, both the wind direction frequency and WS changed slightly with different CTs. Mean WS for different CTs varied within 0.1 m s⁻¹, ranging from 1.42 to 1.51 m s⁻¹. This indicates that WS in Chongqing was likely dominated by local complicated topography. In Chengdu, WS ranged from 1.94 ± 1.04 m s⁻¹ (associated with CT 3) to 2.49 ± 1.46 m s⁻¹ (associated with CT 4). WS in Chengdu was controlled by large-scale circulation and also impacted by surrounding topography, as discussed in previous studies (Ye and Gao, 1979; Wang and Tan, 2014; Feng *et al.*, 2016). The small pressure gradient resulted from the weak high-pressure system could explain the minimum WS in CT 3. High WS associated with CT 4 can be a result of two flows, one northerly flow over Chengdu induced by low-pressure system and another one originated from the high-pressure system to the northwest along the northern and western edges of the basin. Similar to the latter flow, for CT 2 the topography turned the easterly flow into northerly flow with smaller WS over Chengdu. Therefore, in Fig. 4(a) the pattern of wind direction frequency for CT 2 was similar to that for CT 4, but the WS was much smaller especially for the northerly wind. In CT 1 more southerly winds were observed because the strong high-pressure system caused more geostrophic features for the air flow.

Horizontal dispersion was quantified by considering air mass stagnation, recirculation and ventilation following the method proposed by Allwine and Whiteman (1994). The displacement (L) of the air mass in any 24-hour period was calculated from local average wind vector. Similarly, the air moving distance (D) was calculated from the scalar average of WS during the same period. The recirculation

index can be expressed as $R = 1 - L/D$. A value of 1 for R represents no net transport and a value of 0 means constant flow direction. Recirculation and stagnation were identified as $R \geq 0.6$ and $D \leq 130$ km (scalar mean WS is about 1.5 m s⁻¹), respectively. Ventilation was identified as $D \geq 250$ km (with a scalar mean WS of about 2.9 m s⁻¹) and $R \leq 0.2$ (Ye *et al.*, 2016). The occurrence frequencies of stagnation, recirculation and ventilation for each CT in Chengdu and Chongqing are listed in Table 2.

In Chongqing, stagnation occurred frequently while ventilation rarely happened due to calm winds. The relatively lower frequency of stagnation for CT 4 than for other CTs was in accordance with the SEC values in Table 1. In Chengdu, stagnation occurred much less but ventilation occurred more than in Chongqing. In both cities, the highest ventilation and lowest stagnation frequencies were associated with CT 4, which were consistent with the low SEC for this CT. Conversely, the frequent stagnation and no ventilation associated with CT 3 were responsible for the high SEC. At both cities, the frequencies of recirculation were higher for CT 3 than for other CTs due to the weak weather system for CT 3, under which condition local circulations with diurnal features, such as mountain-valley breeze and urban heat island breeze, became dominant.

The patterns of pressure vertical velocities for each CT are presented in Fig. 5. The study area lies at the lee side of the Tibetan Plateau, and thus subsidence features are expected. However, the specific topography in the basin area favors the formation of cyclonic-like circulation in the low-level troposphere, as discussed above. This prevailing circulation pattern induces convergence and further causes ascending flow in the center of the basin area. Hence, strong subsidence was observed around Chengdu and Chongqing (Fig. 5), which are located at the edges of the basin, and there was always an updraft zone between these two cities. Chengdu was in the kernel of the western subsidence zone

Table 1. Averages and standard deviations of SEC and key meteorological parameters for each CT in Chengdu and Chongqing.

CT	Cheng Du					Chong Qing				
	SEC (km ⁻¹)	RH (%)	WS (m s ⁻¹)	PBLH (m)	T (°C)	SEC (km ⁻¹)	RH (%)	WS (m s ⁻¹)	PBLH (m)	T (°C)
1	0.84 ± 0.73 (2, 3, 4)	67.35 ± 19.40 (2, 4)	2.08 ± 1.17 (3, 4)	870.56 ± 351.14 (2, 3, 4)	7.77 ± 3.05 (2, 3, 4)	1.00 ± 0.55 (3, 4)	76.14 ± 11.95 (2, 3)	1.42 ± 0.67 (2, 3, 4)	1040.23 ± 415.81 (2, 4)	8.91 ± 2.52 (2, 3, 4)
2	1.02 ± 0.85 (1, 4)	72.67 ± 13.22 (1, 3, 4)	2.03 ± 1.14 (3, 4)	740.75 ± 282.27 (1, 3, 4)	9.85 ± 3.40 (1, 3, 4)	1.02 ± 0.57 (3, 4)	73.32 ± 12.31 (1, 4)	1.45 ± 0.73 (1)	954.81 ± 372.61 (1)	11.12 ± 3.34 (1, 3, 4)
3	1.09 ± 0.67 (1, 4)	68.36 ± 17.37 (2)	1.94 ± 1.04 (1, 2, 4)	809.78 ± 275.14 (1, 2, 4)	9.15 ± 3.83 (1, 2, 4)	1.10 ± 0.80 (1, 2, 4)	73.33 ± 13.42 (1, 4)	1.47 ± 0.72 (1)	1046.95 ± 371.38 (-)	10.46 ± 3.30 (1, 2, 4)
4	0.73 ± 0.51 (1, 2, 3)	68.98 ± 16.76 (1, 2)	2.49 ± 1.46 (1, 2, 3)	944.52 ± 346.96 (1, 2, 3)	8.18 ± 3.51 (1, 2, 3)	0.89 ± 0.54 (1, 2, 3)	75.56 ± 11.85 (2, 3)	1.51 ± 0.79 (1)	969.80 ± 455.08 (1)	9.29 ± 3.19 (1, 2, 3)
Overall	0.90 ± 0.42	68.99 ± 17.43	2.07 ± 1.22	838.23 ± 329.95	8.56 ± 3.49	1.00 ± 0.61	74.81 ± 12.38	1.46 ± 0.72	1003.46 ± 407.31	9.81 ± 3.16

Note: The numbers in each parenthesis represent the CTs for which the averages were significantly different with the average above the parenthesis at a 0.05 level.

and exhibited strong downdraft while Chongqing was closer to the updraft zone. The strongest and weakest subsidence occurred in CT 3 and CT 4, respectively. Chongqing was even in the updraft zone when CT 4 prevailed. These circulation patterns contributed to the high SEC for CT 3 and low SEC for CT 4 in both cities.

The mean PBLH for each CT in the study area are presented in Fig. 6. Generally, a low PBLH belt existed in the western and southern basin area, which might be caused by the subsidence of lee effect. As discussed above, there was always an ascending region caused by the large-scale circulation and topography. As a result, a high PBLH region extended from northeastern corner to the center of the basin. Chengdu and Chongqing are situated at the edge of low belt and high region, respectively. Hence, the mean PBLH of Chongqing was higher than that in Chengdu as listed in Table 1. The between-CT comparison of SEC and PBLH showed that SEC increases with decreasing PBLH in Chengdu, but no such trend existed in Chongqing. This further implies that the influence of atmospheric circulation was weaker in Chongqing than in Chengdu.

The temperature in Chengdu and Chongqing varied synchronously with different CTs. The strong northerly cold air in CT 1 caused the lowest temperature. The easterly flow in CT 2 brought warmer air mass into the study area, so the highest temperature was associated with CT 2. The temperature in CT 3 was higher than that in CT 4 because there was more advection from the north and more cloud cover in CT 4 (Fig. 5).

The between-CT variations of RH and T in Chengdu were different than those in Chongqing according to Table 1. In Chongqing, the between-CT mean RH and T for different CTs were negatively correlated. This might be related to the fact that Chongqing was less influenced by the large-scale circulation as demonstrated by the other parameters. Hence, the water vapor content in the air should be stable for different CTs. In Chengdu, evidently water vapor advection could be noticed in CT 1 and CT 2. Specifically, severe influence of the northerly cold and dry air mass caused the lowest T and lowest RH in CT 1, and the easterly flow resulted in the highest T and highest RH in CT 2. In terms of between-CT mean, the relations between the SEC and RH, T were complicated in both cities.

Temperature inversion is another factor influencing air quality through suppressing vertical mixing of pollutants. Fig. 7 presents the occurrence frequencies of inversion at different altitudes and with different temperature gradients for each CT. The frequency of inversion was 43.1%, 37.6%, 62.9% and 33.1% in Chengdu, and 10.9%, 17.0%, 35.1% and 8.4% in Chongqing for CT 1, 2, 3 and 4, respectively. Besides the higher frequencies of inversion found in Chengdu than Chongqing, the inversion height was much lower and the inversion strength (temperature gradient) was much higher in Chengdu. The frequency of inversion was directly related to the atmospheric circulation patterns. For example, inversion occurred most frequently in CT 3 because the high-pressure system caused clear weather and facilitated nocturnal radiative cooling, which favored the formation of surface inversion. Besides, the

Fig. 4. The frequency and mean wind speed at different wind direction for the 4 CTs in (a) Chengdu and (b) Chongqing.

Table 2. The occurrence frequency of stagnation, recirculation and ventilation for 4 CTs in Chengdu and Chongqing.

Site	CT	Occurrence Frequency (%)		
		Stagnation	Recirculation	Ventilation
Chengdu	1	15.53	15.53	11.65
	2	13.46	5.77	7.69
	3	16.67	29.17	0
	4	7.23	12.05	32.53
	Overall	13.52	15.29	12.45
Chongqing	1	73.98	20.07	0
	2	71.36	15.91	0.45
	3	70.00	23.57	0
	4	62.20	13.41	0.61
	Overall	69.48	18.35	21.37

Fig. 5. The pressure vertical velocity distributions on the cross section passing through Chengdu and Chongqing for the 4 CTs. The gray areas represent underlying topography. The left and right dashed lines label the locations of Chengdu and Chongqing, respectively.

strong subsidence associated with the high-pressure system led to more upper-level inversion. On the contrary, the low-pressure system and weak subsidence in CT 4 led to the lowest frequency of inversion.

In summary, differences in SEC and meteorological parameters between different CTs were much larger in Chengdu than Chongqing, indicating stronger influence of atmospheric circulation in Chengdu. The synoptic dynamic factors, such as wind, subsidence, inversion and PBLH, dominated the between-CT variations in SEC in both cities.

No apparent impact of the chemistry-related factors (RH and T) was recognized for causing between-CT SEC variations.

Within-CT Correlations between SEC and Meteorological Parameters

The within-CT analysis is capable of capturing the key influencing parameters by diminishing the impact of circulation patterns. In any given circulation pattern, the variations of meteorological parameters are mainly caused by the intensity of dominant weather systems. The correlation

coefficients between the relative anomalies of SEC and meteorological parameters, including RH, WS, PBLH and T, for each CT are listed in Table 3.

In Chengdu, SEC negatively correlated with WS for CT 1, CT 2 and CT 4 due to the effect of horizontal dispersion, and only weakly correlated with WS for CT 3 due to calm

wind and frequent stagnation. Due to the strong subsidence over Chengdu, vertical dispersion of pollutants was suppressed. Only CT 1 and CT 4 showed significant negative correlation between PBLH and SEC. SEC positively correlated with RH for all the CTs, which could be explained by several RH-dependent processes in the atmosphere. For

Fig. 6. The within-CT mean planetary boundary layer height at 06 UTC for each CT. The triangles mark the location of Chengdu (west) and Chongqing (east).

Fig. 7. The occurrence frequency of temperature inversion at different height and with different temperature gradient for each CT in Chengdu and Chongqing. The colored shadings are the frequency for corresponding bins of inversion bottom and temperature gradient at inversion bottom. The parenthesized numbers are the overall frequencies of inversion for each CT.

Table 3. The within-CT correlation coefficients between the relative anomalies of SEC and meteorological parameters for each CT in Chengdu and Chongqing.

CT	Chengdu				Chongqing			
	RH	WS	PBLH	T	RH	WS	PBLH	T
1	0.34	−0.24	−0.16	0.09	0.19	−0.23	−0.11	0.23
2	0.13	−0.36	0.08	0.08	0.35	−0.27	−0.22	0.11
3	0.46	−0.03	−0.04	−0.22	0.34	−0.26	−0.14	−0.03
4	0.18	−0.34	−0.15	0.04	0.17	−0.38	−0.02	0.23
Overall	0.21	−0.27	−0.11	−0.02	0.24	−0.22	−0.08	0.10

Note: The bold black and gray data imply the correlations are significant at level of 0.01 and 0.05, respectively.

example, high RH enhances aqueous-phase reactions of sulfate (Sun *et al.*, 2013) and facilitates the formation of ammonium nitrate (Tai *et al.*, 2010; Seinfeld and Pandis, 2016). High RH also modulates partitioning of semi-volatile species into particulate phase (Hu *et al.*, 2008; Zhang *et al.*, 2015b). SEC correlated weakly and positively with temperature for CT 1, CT 2 and CT 4 and negatively for CT 3. The weak positive correlation could be a competitive effect between strong photochemical reactions and restrained condensation of volatile compounds under high-temperature condition (Tai *et al.*, 2010; Pateraki *et al.*, 2012). The significant negative correlation in CT 3 should be a result of the negative correlation between RH and temperature under the weak weather system. Dispersion is important in modulating SEC in strong weather systems (CTs 1, 2 and 4) while chemical process is more critical in weak systems (CT 3).

Correlation analysis results in Chongqing were similar to those in Chengdu for each individual meteorological parameter, but the within-CT combination of meteorological parameters and between-CT discrepancies differed from each other between the two cities. The impacts of dispersion and chemical processes were equally important for every CT in Chongqing due to weak influence of large-scale circulation. Specifically, the significant influence of T on SEC in strong weather systems (CT 1 and CT 4) and WS/PBLH in weak weather systems (CT 3) found in Chongqing was not found in Chengdu.

Interannual Variation of SEC and Meteorological Parameters

Fig. 8 presents the interannual variations of CT occurrence days (F_{nm}) in winters of 2001–2016. The ranges of occurrence days for CTs 1–4 were 16–49, 12–27, 4–29, and 9–32 days, respectively. The interannual variations of SEC and meteorological parameters are shown in Fig. 9 for Chengdu and in Fig. 10 for Chongqing. Seasonal mean SEC in Chengdu first increased before 2005 and then decreased starting in 2006. The largest increase occurred in 2005 with SEC being 0.583 km^{-1} higher than that in 2004, and then a steep decrease of -0.488 km^{-1} happened in 2006, followed by a gradual decrease until 2015 and an accelerated decrease in 2016 (by -0.301 km^{-1} compared to that in 2015) (Table 6). The variation trends of seasonal mean SEC showed an increase of 2.745 km^{-1} per decade during 2001–2005 and a decrease of -0.260 km^{-1} per decade during 2005–2016 (Table 7). Seasonal mean SEC for CT 1,

CT 2 and CT 4 showed similar trends to the overall mean SEC, while that for CT 3 showed a distinct increasing trend during 2009–2012. The variation ranges of SEC were much larger for CT 2 and CT 3 than CT 1 and CT 4.

Different interannual variations were observed in Chongqing than in Chengdu. The whole study period could be divided into 3 intervals: a declining one from 2001 to 2009, an increasing one from 2009 to 2014, and then a declining one again from 2014 to 2016, resulting the lowest seasonal mean SEC in 2009 and the highest in 2014. The magnitudes of interannual variations were much lower than those in Chengdu. The maximum interannual increase and decrease of SEC were 0.176 and -0.162 km^{-1} , respectively. The decreasing trends were -0.338 and -1.297 km^{-1} per decade during 2001–2009 and 2014–2016, respectively, while the increasing trend during 2009–2014 was 0.758 km^{-1} per decade. The between-CT differences of interannual variations were much smaller than those in Chengdu.

Compared with SEC, the overall interannual variations of meteorological parameters were more monotonous in the whole study period. In both Chengdu and Chongqing RH showed a declining trend. WS retained an increasing trend in Chengdu and a decreasing one in Chongqing. PBLH increased before 2007 and then slightly decreased at both cities. The interannual variations of temperature in Chongqing were almost the same as those in Chengdu but with larger magnitudes. Temperature in the study area seemed to be stable during 2001–2016 despite significant fluctuations. The differences of the interannual variation trends between CTs were small for RH and temperature, but significant for WS and PBLH.

The relationships between the variation trends of SEC and meteorological parameters were complicated because of additional dominant influences of non-synoptic factors (such as anthropogenic emissions). As an example, the decreasing RH and increasing PBLH during 2001–2004 in Chongqing can lead to decreasing SEC, while the decreasing WS favors increasing SEC. The variation trends of SEC cannot be explained simply by the trends of meteorological parameters. Further analysis results are presented in the next section.

Synoptic and Non-synoptic Signals and their Variation Trends

The results of the principal component analysis on the relative anomalies of RH, WS, PBLH and T are listed in Table 4. For the i^{th} PC of the m^{th} CT, the loading of each

meteorological parameter ($\sqrt{\lambda_{mi}e_{mi}}$) and the explained variance ($\lambda_{mi} / \sum_{i=1}^4 \lambda_{mi}$) are presented. Each PC represents certain weather condition, described by the combination of

meteorological parameters. A higher loading implies higher impact from the corresponding meteorological parameter. The explained variance of each PC can be interpreted as the occurrence frequency of the corresponding weather condition. Hence, the first PC of each CT stands for the

Fig. 8. The interannual variations of occurrence days in winters of 2001–2016 for each CT.

Fig. 9. The interannual variations of seasonal mean of (a) SEC, (b) RH, (c) WS, (d) PBLH, and (e) T in winters of 2001–2016 for each CT in Chengdu. The dashed lines are the averages for separate CTs. The gray lines are the overall averages.

Fig. 10. The same as Fig. 9 but in Chongqing.

Table 4. The loading of meteorological parameters and explained variance (EV) of each PC for each CT in Chengdu and Chongqing.

CT	PC	Chengdu					Chongqing				
		RH	WS	PBLH	T	EV (%)	RH	WS	PBLH	T	EV (%)
CT1	PC1	0.744	−0.674	−0.590	0.059	33.96	−0.837	0.228	0.803	0.165	35.61
	PC2	0.152	−0.268	0.585	0.860	29.43	−0.032	0.731	−0.092	−0.724	26.72
	PC3	0.505	0.686	−0.125	0.210	19.63	−0.027	−0.625	0.287	−0.667	22.97
	PC4	0.411	−0.062	0.543	−0.461	16.99	0.546	0.148	0.514	0.060	14.70
CT2	PC1	−0.746	0.076	0.734	0.740	41.20	−0.788	0.523	0.644	0.460	38.04
	PC2	0.033	0.942	−0.344	0.278	27.12	−0.180	−0.393	0.607	−0.710	26.49
	PC3	0.656	−0.055	0.250	0.419	16.79	0.286	0.754	0.055	−0.443	21.25
	PC4	0.110	0.321	0.530	−0.447	14.89	−0.514	0.055	−0.463	−0.296	14.22
CT3	PC1	−0.578	0.447	0.773	0.797	44.15	−0.824	0.630	0.476	0.483	38.37
	PC2	0.513	0.818	−0.108	0.018	23.58	0.148	0.521	−0.797	0.360	26.44
	PC3	0.635	−0.353	0.390	0.280	18.94	−0.199	0.437	−0.112	−0.798	21.98
	PC4	−0.017	0.087	0.489	−0.535	13.33	0.510	0.377	0.354	0.029	13.21
CT4	PC1	−0.707	0.133	0.819	0.371	33.15	0.850	−0.293	−0.735	−0.271	35.52
	PC2	0.423	0.661	−0.046	0.673	26.75	0.053	−0.868	0.494	−0.233	26.37
	PC3	0.257	−0.739	0.084	0.570	23.58	0.124	−0.290	−0.084	0.932	24.39
	PC4	−0.505	−0.014	−0.566	0.292	16.52	0.510	0.276	0.458	0.059	13.72

main feature of the weather controlled by this circulation pattern. In Chengdu the first PC of CT 1 was characterized as low RH accompanied by high WS and PBLH, which

was related to the northerly air mass invasion. This weather type should lead to positive correlations between RH and T because of the dry cold air from the north. The

weak positive contribution of T in the first PC of CT 1 might be resulted by the counteraction from local radiative warming. The first PCs of the other CTs in Chengdu and all the CTs in Chongqing showed more localized characteristics, namely weaker WS influences and low RH accompanied by high PBLH and T. The second, third and fourth PCs represent less frequent weather conditions, but even the lowest one explained more than 10% of the variances of these PCs, and thus was non-negligible.

The regression expressions between SEC and PCs are presented in Table 5. The regression slopes are controlled by the correlations between SEC and the dominant meteorological parameters of this PC. Hence, the results in Table 5 need to be in accordance with Tables 3 and 4. For instance, the first PC of CT 1 in Chengdu was dominated by RH, WS and PBLH with positive loading for RH and negative loadings for WS and PBLH. Meanwhile the SEC was positively (negatively) correlated to RH (WS and PBLH) according to Table 3. As a result, the first PC of CT 1 in Chengdu was positively correlated to SEC. The first PC of CT 2 in Chengdu was dominated by RH, PBLH and T with loadings of similar magnitudes. However, the correlations between SEC and these 3 parameters were weak, enabling this PC having little impact on the relative anomalies of SEC. While in the case of the third PC of CT 1 in Chengdu, the opposite influences of RH and WS led to weak correlations between SEC and this PC.

Applying the method described in Sec. 2.4, the synoptic occurrence signals, overall synoptic signals and derived non-synoptic signals were calculated and are presented in Fig. 11. The interannual variations of the observed signals ($\Delta\overline{\text{SEC}}_n$), the synoptic occurrence signals (ΔO_n), and the overall synoptic signals (ΔS_n) are listed in Table 6, along with the ratio $\Delta\text{S}_n / \Delta\overline{\text{SEC}}_n$, which is used to represent the contribution of atmospheric circulation to the SEC

variations. In Chengdu, $\Delta\overline{\text{SEC}}_n$ fell into a wide range, from nearly 0 to 0.583 km^{-1} . By contrast, ΔS_n was much smaller, between 0.010 and 0.077 km^{-1} . In 8 of the 15 years, ΔS_n and $\Delta\overline{\text{SEC}}_n$ had the same sign, which means the atmospheric circulation intensified the interannual variations of SEC, while in the other 7 years the impact of the atmospheric circulation was weakening. The medians of $\Delta\text{S}_n / \Delta\overline{\text{SEC}}_n$ in intensifying and weakening years were 23.17% and 44.79%, respectively. A comparison between ΔO_n and ΔS_n showed that in most years the occurrence and intensity of the circulation had opposite impacts on SEC. In Chongqing, both $\Delta\overline{\text{SEC}}_n$ and ΔS_n varied in smaller ranges than those in Chengdu. The maximum magnitudes of $\Delta\overline{\text{SEC}}_n$ and ΔS_n were 0.176 and 0.062 km^{-1} , respectively. In more years (10 of 15) the contribution of synoptic was positive. The medians of $\Delta\text{S}_n / \Delta\overline{\text{SEC}}_n$ in intensifying and weakening years were 23.58% and -8.02% , respectively, in Chongqing. This quantitative analysis showed that in more than half of the years the impact of the atmospheric circulation intensified the interannual variations of SEC. The medians of the intensifying rate were around 23% in both Chengdu and Chongqing. Hence, the derived non-synoptic signals were generally smoother than the observed ones. For example, the valley value of SEC in 2012 was eliminated after subtracting the influence of the atmospheric circulation.

Table 7 presents the variation trends of $\overline{\text{SEC}}_n$, E_n , O_n and S_n during the periods of 2001–2005 and 2005–2015 in Chengdu and 2001–2009, 2009–2014 and 2014–2016 in Chongqing. The ratios of the trends of E_n , O_n and S_n to those of $\overline{\text{SEC}}_n$ are also listed to show the contributions of these signals to the trends of SEC. In Chengdu, SEC increased (decreased) during 2001–2005 (2005–2015) with a trend of 2.745 (-0.188) km^{-1} per decade. During the growth period, introducing the intensity of circulation counteracted the effect of occurrence frequency. Only 0.15% of the SEC

Table 5. The SEC-PC regression expressions and corresponding correlation coefficients between the relative anomalies of SEC and PCs for each CT in Chengdu and Chongqing.

CT	Chengdu		Chongqing	
	regression	R	regression	R
CT1	$0.653 \times \text{PC1} + 0.029$	0.83	$-0.200 \times \text{PC1} + 0.002$	-0.73
	$0.221 \times \text{PC2} - 0.020$	0.60	$-0.468 \times \text{PC2} - 0.032$	-0.81
	$-0.040 \times \text{PC3} - 0.029$	-0.12	$-0.215 \times \text{PC3} - 0.047$	-0.58
	$0.139 \times \text{PC4} - 0.022$	0.28	$-0.171 \times \text{PC4} - 0.008$	0.82
CT2	-0.028	-	$-0.475 \times \text{PC1} + 0.019$	-0.82
	$-0.375 \times \text{PC2} + 0.023$	-0.55	$-0.359 \times \text{PC2} - 0.021$	-0.76
	$0.463 \times \text{PC3} + 0.021$	0.95	$-0.506 \times \text{PC3} + 0.019$	-0.90
	$-0.256 \times \text{PC4} - 0.007$	-0.45	$-0.117 \times \text{PC4} - 0.014$	-0.42
CT3	$-0.053 \times \text{PC1} + 0.022$	-0.12	$-0.743 \times \text{PC1} - 0.015$	-0.89
	$0.019 \times \text{PC2} + 0.002$	0.04	$0.157 \times \text{PC2} - 0.019$	0.27
	$-0.049 \times \text{PC3} - 0.010$	-0.11	$-0.234 \times \text{PC3} - 0.029$	-0.49
	$0.316 \times \text{PC4} - 0.029$	0.46	$-0.383 \times \text{PC4} + 0.002$	-0.85
CT4	$-0.268 \times \text{PC1} + 0.026$	-0.73	$-0.003 \times \text{PC1} - 0.025$	-0.01
	$-0.312 \times \text{PC2} - 0.015$	-0.69	$0.310 \times \text{PC2} - 0.010$	0.68
	$0.361 \times \text{PC3} + 0.012$	0.84	$0.684 \times \text{PC3} + 0.052$	0.91
	$0.151 \times \text{PC4} + 0.035$	0.46	$-0.295 \times \text{PC4} - 0.009$	-0.61

Note: The bold numbers in black and gray indicate the correlations are significant at level of 0.01 and 0.05, respectively.

Fig. 11. The interannual variations of $\overline{\text{SEC}}_n$, E_n , O_n and S_n in Chengdu and Chongqing.

Table 6. The interannual variations of $\overline{\text{SEC}}_n$, O_n and S_n in unit of km^{-1} , and the ratios of the variations of S_n to those of $\overline{\text{SEC}}_n$ in Chengdu and Chongqing.

year	Chengdu				Chongqing			
	$\Delta\overline{\text{SEC}}_n$ (km^{-1})	ΔO_n (km^{-1})	ΔS_n (km^{-1})	$\frac{\Delta S_n}{\Delta\overline{\text{SEC}}_n}$ (%)	$\Delta\overline{\text{SEC}}_n$ (km^{-1})	ΔO_n (km^{-1})	ΔS_n (km^{-1})	$\frac{\Delta S_n}{\Delta\overline{\text{SEC}}_n}$ (%)
2002–2001	0.098	−0.037	−0.028	−28.82	0.106	−0.009	0.019	18.09
2003–2002	0.147	0.036	0.031	20.81	0.069	0.009	−0.025	−35.42
2004–2003	0.314	−0.025	−0.019	−5.95	−0.125	−0.004	0.001	−0.53
2005–2004	0.583	0.027	0.013	2.17	0.006	−0.001	0.009	140.74
2006–2005	−0.488	−0.026	−0.050	10.20	−0.068	−0.006	−0.023	34.03
2007–2006	−0.015	−0.001	0.011	−74.35	0.035	0.006	−0.001	−2.20
2008–2007	0.012	−0.030	−0.038	−315.67	−0.143	−0.019	−0.021	14.89
2009–2008	0.102	0.052	0.051	50.19	−0.130	0.027	0.045	−35.12
2010–2009	−0.076	0.017	0.010	−13.51	0.159	0.005	−0.013	−8.02
2011–2010	0.069	−0.043	−0.031	−44.79	0.058	−0.018	0.025	43.63
2012–2011	−0.169	−0.054	−0.043	25.52	−0.066	−0.027	−0.062	94.44
2013–2012	0.100	0.084	0.077	76.82	0.138	0.034	0.040	29.07
2014–2013	−0.001	−0.014	−0.019	1716.48	0.176	0.000	0.024	13.44
2015–2014	0.043	−0.020	−0.024	−55.18	−0.097	−0.003	−0.005	5.45
2016–2015	−0.301	0.008	−0.014	4.53	−0.162	−0.001	−0.007	4.22

Note: The shaded number means the overall synoptic signal varies in opposite direction to the observed signal in this year.

trend was attributed to atmospheric circulation, which elucidates that the strong increase was the result of non-synoptic factors. During the slight decline period the contribution of atmospheric circulation accounted for 7.96% of the decreasing trend. SEC in Chongqing decreased, increased and then decreased again during 2001–2009, 2009–2014 and 2014–2016 with trends of $−0.338$, 0.758 and $−1.297 \text{ km}^{-1}$ per decade, respectively. Similar to Chengdu, the contributions of atmospheric circulation were relatively higher during the declining periods. During 2001–2009 and 2014–2016, about 10.00% and 4.68%,

respectively, of the increasing trends were caused by atmospheric circulation. By contrast, the contribution during 2009–2014 was 0.67%.

CONCLUSIONS

In this study, the characteristics of atmospheric circulation in the Sichuan Basin during the winters of 2001–2016 were first analyzed by CTC. Then, based on the results, the effect of circulation patterns on the relationship between the SEC and meteorological parameters was

Table 7. The variation trends of $\overline{\text{SEC}}_n$, E_n , O_n and S_n in unit of km^{-1} per decade, and the ratios of the trends of E_n , O_n and S_n to those of $\overline{\text{SEC}}_n$ during selected periods in Chengdu and Chongqing.

Signal	Chengdu (km^{-1} per decade)		Chongqing (km^{-1} per decade)		
	2001–2005	2005–2015	2001–2009	2009–2014	2014–2016
$\overline{\text{SEC}}_n$	2.745	−0.188	−0.338	0.758	−1.297
E_n	2.741	−0.173	−0.305	0.753	−1.236
	99.85%	92.04%	90.00%	99.33%	95.32%
O_n	0.012	−0.011	−0.010	−0.026	−0.018
	0.45%	5.99%	2.81%	−3.39%	1.39%
S_n	0.004	−0.015	−0.034	0.005	−0.061
	0.15%	7.96%	10.00%	0.67%	4.68%

investigated. In both Chengdu and Chongqing, a weak high-pressure system (CT 3) and a local low-pressure system (CT 4) were responsible for the highest and lowest SEC values, respectively, whereas the northerly centered high (CT 1)- and low (CT 2)-pressure systems were associated with medium-level air pollution. Synoptic dynamic factors, such as wind, subsidence, inversion and PBLH, dominated the variation in SEC between CTs. Furthermore, the influence of atmospheric circulation was stronger in Chengdu than Chongqing.

A new method for discriminating synoptic from non-synoptic signals in the observed SEC series was developed to assess the contributions of synoptic signals to the interannual variations of the SEC. This method improved upon earlier studies, which used a single presumed parameter, by applying multiple influential meteorological parameters to assess the effect of atmospheric circulation intensity on the SEC. The interannual SEC variations in these cities were heightened by atmospheric circulation in more than half of the years but reduced by it in the remaining years. Additionally, circulation contributed 5–10% to the decreasing SEC trend but less than 1% to the increasing trend.

The selection of influential parameters is critical in this newly developed method and usually based on a priori knowledge. In addition to the four parameters (WS, RH, T and PBLH) considered in this work, factors such as wind direction, atmospheric inversion and vertical velocity may also significantly influence the SEC. Since these factors are directional or related to vertical profiles, they must be parameterized, which requires further research.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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