Supplementary materials for

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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S1. Evaluation of relative errors of SEC caused by hygroscopic growth correction

We investigated the aerosol composition information in the study area, and estimated the hygroscopic growth correction factors using a revised IMPROVE algorithm. By comparing these correction factors with those used in this study, we can give the range of the errors caused by hygroscopic growth correction.

The composition of wintertime aerosol in Sichuan Basin were seldomly reported. Yang et al. (2011) and Wang et al. (2004) gave the $PM_{2.5}$ composition in winter of 2002 in Chengdu. Wang et al. (2018) reported the $PM_{2.5}$ composition in 2014 and 2015. The composition data were summarized in Table S-1. The mass proportion of $(NH_4)_2SO_4$ and NH_4NO_3 were obtained by allocating NH_4^+ to SO_4^{2-} and NO_3^-.

Table S-1. aerosol composition in Chengdu from literatures.

year	SO_4^{2-}	NO_3^-	NH_4^+	OC	EC	$(NH_4)_2SO_4$	NH_4NO_3
2002	14.23%	5.32%	4.68%	29.10%	5.32%	17.86%	6.37%
2014-2015	14.45%	15.42%	11.19%	17.36%	5.55%	20.58%	20.48%

Wang et al. (2017) used a revised IMPROVE algorithm (Marc et al., 2007) to calculate the aerosol scattering coefficients, and compared these computed scattering coefficients to observations. They concluded that the average relative error between the estimated and measured scattering coefficient was 3% for the revised IMPROVE algorithm. According to Wang et al. (2017), the dominant contributors for the atmospheric extinction were $(NH_4)_2SO_4$, NH_4NO_3, organic matters (OM) and elemental carbon (EC), accounting 90% of the atmospheric extinction in winter in Chengdu. We set the contribution of the other factors, including coarse particles, fine soil, Rayleigh scattering and molecular absorption, to be 10%. And the $PM_{2.5}$ mass concentration was set to be 100 μg/m^3. Implementing the revised IMPROVE algorithm, the calculated hygroscopic growth correction factors were shown in the Fig. S-1.

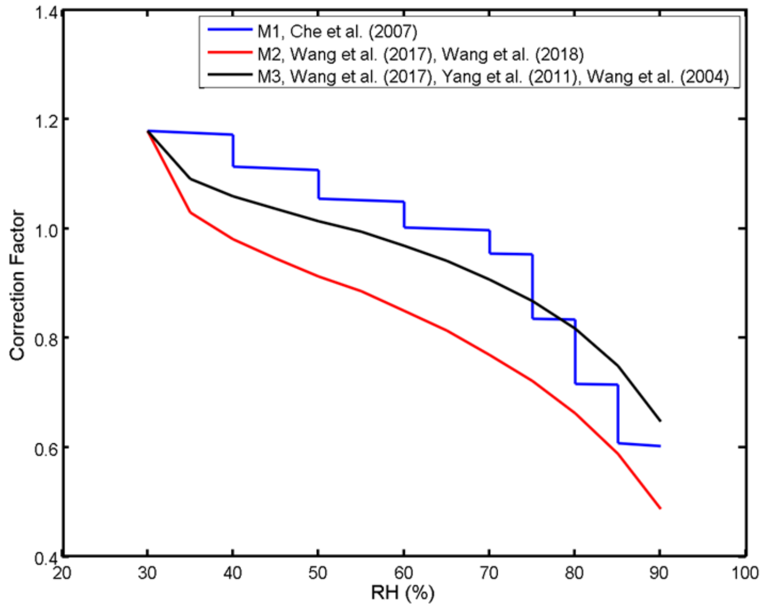

Fig. S-1. The hygroscopic growth correction factors for different aerosol composition

Here, M1 represents the correction factors used in this study, M2 (M3) represents the computed correction factors using the composition data in 2014-2015 (2002). The large discrepancies between M2 and M3 indicate the significant impact of aerosol composition on hygroscopic growth correction factors. The correction factors of M1 are close to those of M3. Hence, the correction method in this study is relatively more appropriate in the starting years of the study period (2001-2016). The lack of the composition information between 2002-2014 makes the estimation of correction factors in this period impossible. Therefore, we kept using the correction factors of M1 in the whole study period. The correction factors of M2 could be used as extreme values to estimate the upper limit of SEC errors.

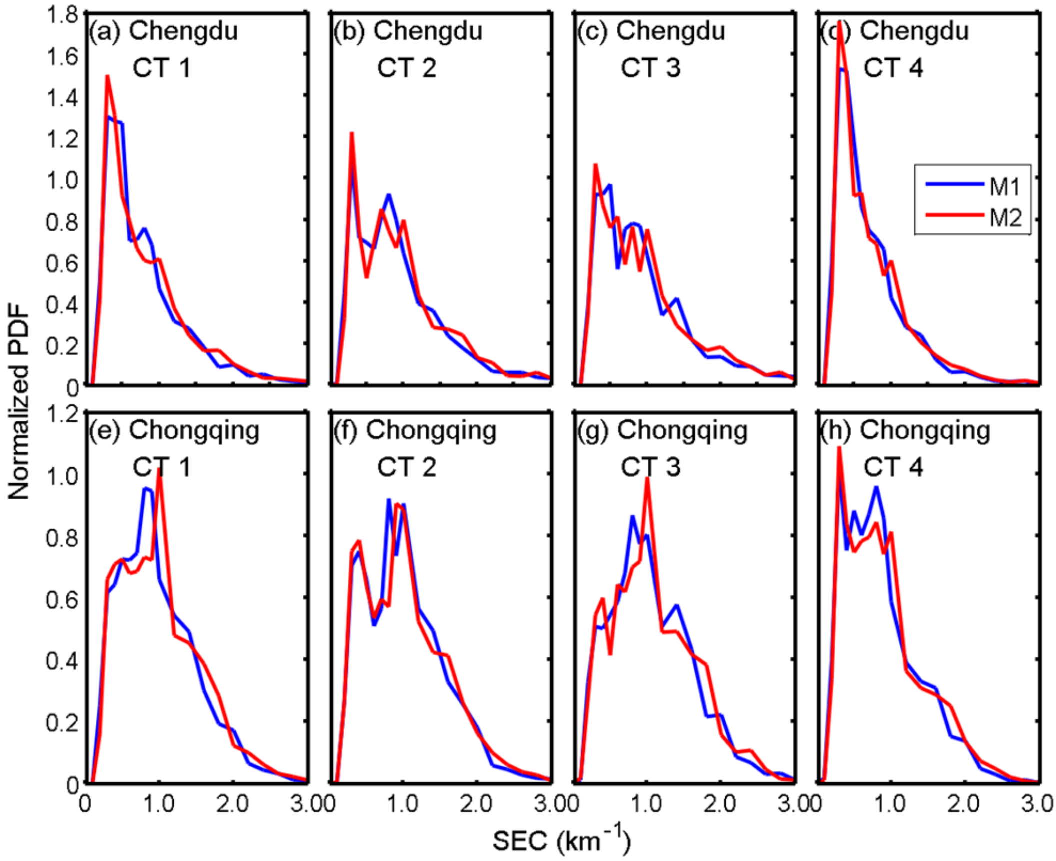

Fig. S-2. The normalized PDF of SEC calculated using different hygroscopic growth correction factors

The normalized probability distribution functions (PDF) of SEC calculated using correction factors of M1 and M2 are presented in Fig. S-2. And the mean differences between SEC from M1 and M2 are listed in Table S-2. In Chengdu, the relative errors caused by hygroscopic growth correction ranges from 12.49% (CT4) to 24.75% (CT3). The relative errors are smaller in Chongqing (around 11%). It is worth to note that these errors represent the upper limit.

Table S-2. The upper limits of relative SEC errors caused by hygroscopic growth correction.

Site	CT 1	CT 2	CT 3	CT 4
Chengdu	0.13 (15.45%)	0.18 (17.23%)	0.27 (24.75%)	0.09 (12.49%)
Chongqing	0.11 (10.87%)	0.11 (11.01%)	0.13 (11.46%)	0.10 (11.61%)

S2. The main steps of calculating ΔSEC_{nm} term

The main steps of calculating ΔSEC_{nm} term are listed below.

In step 1, for each m-th CT, Eq. (1) was used to calculate the relative anomalies for the daily averages of SEC ($\boldsymbol{V}_{SEC,m}$), RH ($\boldsymbol{V}_{RH,m}$), WS ($\boldsymbol{V}_{WS,m}$), PBLH ($\boldsymbol{V}_{PBLH,m}$) and T ($\boldsymbol{V}_{T,m}$) and the matrix $\mathbf{M}_m=[\boldsymbol{V}_{RH,m} \quad \boldsymbol{V}_{WS,m} \quad \boldsymbol{V}_{PBLH,m} \quad \boldsymbol{V}_{T,m}]$ was then constructed.

In step 2, PCA was conducted for the matrix $\mathbf{M}_m$ to find the principal components ($\boldsymbol{PC}_{mi}$, i=1, …, 4) of multiple meteorological parameters. Several techniques have been used to quantify the influence of multiple meteorological parameters on air quality, such as generalized additive models and self-organizing maps (Pearce et al., 2011a; Pearce et al., 2011b). In principal, these methods were based on multi-dimension regression, and thus hard to be interpreted physically. By contrast, each component obtained by the PCA technique can be easily interpreted as certain combination of meteorological parameters. The PCA was conducted by finding the eigenvectors and eigenvalues of correlation matrix. To eliminate the influence caused by the differences of the variations scales between variables, each column of $\mathbf{M}_m$ is normalized by its standard deviation ($\sigma_{X,m}$, X is RH, WS, PBLH or T) and then the 4×4 dimensional correlation matrix $\mathbf{R}_m$ corresponding to this normalized matrix was calculated. By implementing the eigenvalue decomposition, we can find the eigenvalues (λ_{mi}, i=1,…,4) and eigenvectors ($\boldsymbol{e}^*_{mi}$, i=1, …, 4) of $\mathbf{R}_m$. The 4 elements of $\boldsymbol{e}^*_{mi}$ stand for the contributions of RH, WS, PBLH and T, respectively. Each element of $\boldsymbol{e}^*_{mi}$ was then divided by the corresponding $\sigma_{X,m}$. This new vector was scaled by its length to get the unit vector $\boldsymbol{e}_{mi}$. Finally, the daily series of $\boldsymbol{PC}_{mi}$ were calculated using:

$$[\boldsymbol{PC}_{m1} \quad \boldsymbol{PC}_{m2} \quad \boldsymbol{PC}_{m3} \quad \boldsymbol{PC}_{m4}]=\mathbf{M}_m[\boldsymbol{e}_{m1} \quad \boldsymbol{e}_{m2} \quad \boldsymbol{e}_{m3} \quad \boldsymbol{e}_{m4}] \qquad \text{(S-1)}$$

In step 3, for each $\boldsymbol{PC}_{mi}$, the relationships between the relative anomalies of SEC ($\boldsymbol{V}_{SEC,mi}$) and $\boldsymbol{PC}_{mi}$ were established by linear fitting. In order to reduce the noises caused by other $\boldsymbol{PC}$s, $\boldsymbol{V}_{SEC,mi}$ was grouped into 21 bins according to $\boldsymbol{PC}_{mi}$ varying from -1 to 1 with a step of 0.1. $\boldsymbol{V}_{SEC,mi}$ was averaged within each group and linearly fitted with the binned $\boldsymbol{PC}_{mi}$ to obtain the relationships between $\boldsymbol{V}_{SEC,mi}$ and $\boldsymbol{PC}_{mi}$ according to:

$$\boldsymbol{V}_{SEC,mi}=A_{mi}\boldsymbol{PC}_{mi}+B_{mi} \qquad \text{(S-2)}$$

In step 4, ΔSEC_{nm} was calculated by implementing the fitting Eq. (S-2) to the seasonal mean time series. Due to the orthogonality of PCA, the principal components are mutually independent for each CT. Therefore, the derived SEC anomalies from these principal components are additive. The overall anomalies of SEC can be expressed as:

$$\Delta SEC_{nm}=\overline{SEC_m}\cdot\sum_{i=1}^{4}[\lambda_i(A_{mi}\boldsymbol{PC}_{nmi}+B_{mi})] \qquad \text{(S-3)}$$

where $\boldsymbol{PC}_{nmi}$ is the i-th principal component for the relative anomalies of the seasonal mean meteorological parameters in year n for the m-th CT:

$$\boldsymbol{PC}_{nmi}=\left[\overline{\boldsymbol{V}_{RH,nm}} \quad \overline{\boldsymbol{V}_{WS,nm}} \quad \overline{\boldsymbol{V}_{PBLH,nm}} \quad \overline{\boldsymbol{V}_{T,nm}}\right]\boldsymbol{e}_{mi} \qquad \text{(S-4)}$$

$$\overline{\boldsymbol{V}_{X,nm}}=\frac{\overline{X_{nm}}-\overline{X_m}}{\overline{X_m}},\ X=RH, WS, PBLH, or\ T \qquad \text{(S-5)}$$

Here, $\overline{X_m}$ is the 17-year mean of parameter X for the m-th CT.

S3. The probability distribution of SEC

The normalized PDF of SEC for each CT were shown in Fig. S-3. The area under each curve was normalized to be 1. The shapes of these curves agree with the t-test results in Table 1. Especially, CT 4 hold more low SEC, while CT 2 and CT 3 showed more high SEC.

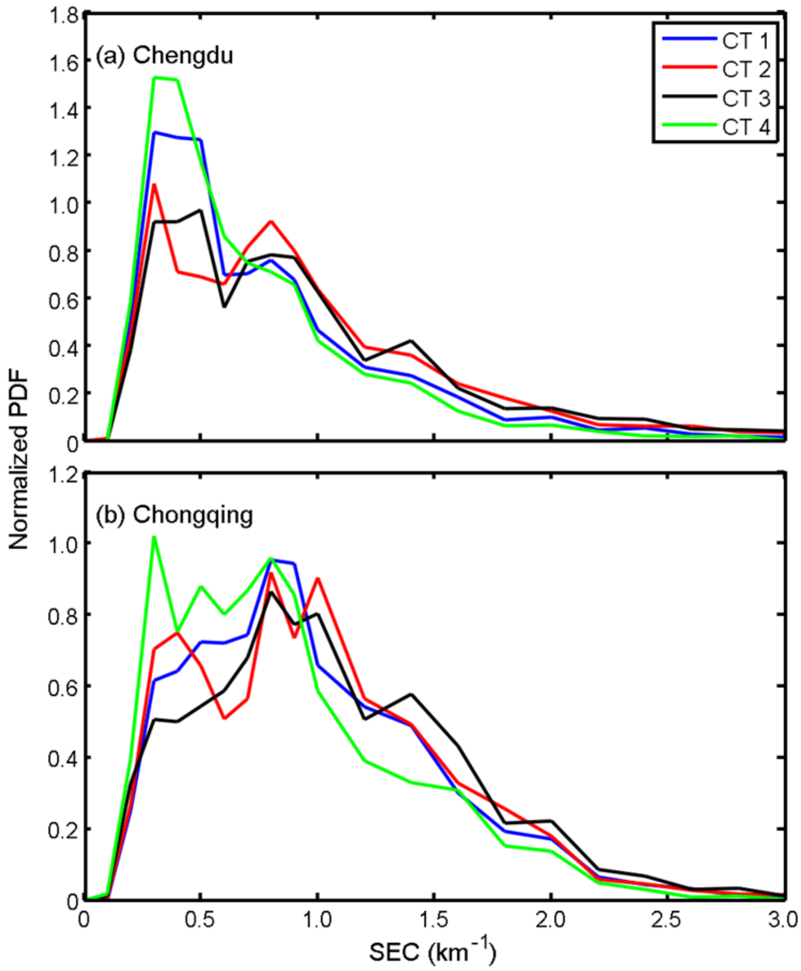

Fig. S-3. The normalized PDF of SEC

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