A Comparison of Meteorology-Driven Interannual Variations of Surface Aerosol Concentrations in the Eastern United States, Eastern China, and Europe

Jin Feng[1, 2], Hong Liao[3] and Yixuan Gu[1, 2]

[1]*State key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS), Beijing, China*

[2]*University of Chinese Academy of Sciences (UCAS), Beijing, China*

[3]*School of Environmental Science and Engineering, Nanjing University of Information Science & Technology (NUIST), Nanjing, China*

Abstract

By the GEOS-Chem simulation with fixed anthropogenic and biomass burning emissions, this study exhibits the differences in interannual variations (IAVs) of surface-layer $PM_{2.5}$ concentrations among three populated regions, the eastern United States (US), eastern China, and Europe driven by variations in meteorological parameters. In the eastern US, $PM_{2.5}$ concentrations have relatively small IAVs with no explicit seasonality, with the absolute percent departure from the mean (APDM) values of 4−16% in four seasons. The IAVs of $PM_{2.5}$ are found to be large in North China and the northwestern Europe during winter and spring. The APDM values are 24−28% in winter and 32−36% in spring in eastern China, and 32−36% in winter and 20−24% in spring in Europe. Additionally, we obtain the key meteorological parameters that drive the IAVs of $PM_{2.5}$ by the stepwise multiline regression model (SLR) containing 8 meteorological variables. The most important meteorological variables over the eastern US, eastern China, and Europe are, respectively, the westerly at 850 hPa, surface wind speed, and the planetary boundary layer height in winter, and precipitation, relative humidity, and surface temperature in summer.

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1. Introduction

Aerosols are major air pollutants that have adverse effects on human health, reduce atmospheric visibility, and influence global climate change. Observational and modeling studies have reported high aerosol concentrations in the eastern United States (US) (Malm 2004; Park et al. 2003, 2004; Walker et al. 2012), eastern China (Zhang et al. 2012b; Wang et al. 2013; Mu and Liao 2014), and Europe (Megaritis et al. 2014; Andersson et al. 2007) as a result of the large anthropogenic emissions of aerosols and aerosol precursors in these regions (Dutkiewicz et al. 2000; Vestreng et al. 2007; Hand et al. 2012; Mijling et al. 2013). Previous studies also showed that the interannual variations (IAVs) of aerosols were significant in the eastern US (Alston et al. 2012), eastern China (Yang et al. 2011; Mu and Liao 2014), and Europe (Andersson et al. 2007). The IAVs in aerosol concentrations were found to be sensitive to the IAVs in anthropogenic emissions of aerosols and aerosol precursors (Mylona 1996; Irie 2005; Ohara et al. 2007; Vestreng et al. 2007; Xing et al. 2013) and in meteorological conditions, since the meteorology modulates the processes of dilution, transport, deposition, and chemical reaction (Andersson et al. 2007; Zhao et al. 2012; Mu and Liao 2014).

Previous studies on the IAVs of aerosol concentrations were insufficient in two aspects: (1) these studies on the IAVs of aerosols were focused on one of the three regions, hardly compared the differences among the regions and seasons due to various dominant meteorological variables; (2) many studies were focused on the net effect of all meteorological variables on the IAVs of aerosols, other than the relative importance of different meteorological variables including temperature, precipitation, humidity, and surface wind speed. This study presents a comparison of meteorology-driven IAVs of seasonal mean $PM_{2.5}$ in the eastern US, eastern China, and Europe by the simulations of year 1986−2006 aerosol concentrations using the global chemical transport model (CTM) GEOS-Chem. Our aims are to quantify the differences in the geographic distributions of the IAVs of $PM_{2.5}$ and to obtain the key meteorological variables that drive the IAVs of aerosols in the eastern US, eastern China and Europe in different seasons.

2. GEOS-Chem simulation

With the GEOS-Chem model (http://acmg.seas.harvard.edu/geos), driven by the assimilated meteorological data from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO), we simulate global aerosols over 1986−2006 with a horizontal resolution of 2° × 2.5° and a vertical resolution of 30 hybrid pressure-sigma layers from the surface to 0.01 hPa. The GEOS-4 meteorological fields, which have long time coverage for 1986−2006, are used because our study needs decades of simulation to get enough samples for interannual variation. The GEOS-Chem model has a fully coupled treatment of tropospheric ozone-NO_x-VOC-aerosol chemistry including various aerosols of sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+) (Park et al. 2004), OC, BC (Park et al. 2003), mineral dust (Fairlie et al. 2007), and sea salt (Alexander et al. 2005; Jaeglé et al. 2011). The model uses the advection scheme of Lin and Rood (1996), the deep convective scheme of Zhang and McFarlane (1995), the shallow convection scheme of Hack (1994), the wet deposition scheme of Liu et al. (2001), and the dry deposition scheme of Wesely (1989) and Wang (1998). The simulated $PM_{2.5}$ is reconstructed by the sum of SO_4^{2-}, NO_3^-, NH_4^+, BC and OC, while mineral dust and sea salt are omitted because they are not the dominant components of $PM_{2.5}$ in the eastern US (Eldred et al. 1997; Malm 2004), eastern China (Ye 2003; Duan et al. 2006), and Europe (Querol et al. 2004). We use the anthropogenic emission inventories of EPA/NEI99 (http://www.epa.gov/ttn/chief/net/1999inventory.html) in the US, Streets et al. (2003) in China, and EMEP (http://www.ceip.at/) in Europe. We fix anthropogenic emissions to the levels of year 2005 in our simulation of year 1986−2006 aerosols in order to highlight the effects of meteorology. The biomass burning emission is also fixed although it does not play a major role in the IAVs of aerosols in these three regions (Voulgarakis et al. 2015). In our simulation, the multi-annual mean $PM_{2.5}$ concentrations over 1986−2006 in the eastern US (95°W−72°W, 30°N−45°N), eastern China (110°E−125°E, 20°N−42°N) and Europe (5°W−30°E, 40°N−60°N) were higher than in

Corresponding author: Hong Liao, School of Environmental Science and Engineering, NUIST, Nanjing, China. E-mail: hongliao@nuist.edu.cn.

other regions in the northern mid-latitudes in December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), and September-October-November (SON) (see supplementary material, Fig. S1). The magnitude and spatial distribution of simulated aerosols in the GEOS-Chem model have been evaluated extensively in previous studies for the US (Park et al. 2003, 2004, 2006; Heald et al. 2006; van Donkelaar et al. 2006; Fu et al. 2009; Drury et al. 2010; Leibensperger et al. 2011; Zhang et al. 2012a), China (Zhang et al. 2012b; Wang et al. 2013) and Europe (Leibensperger et al. 2011; Ma and Yu 2014) for different years and seasons between 1998 and 2008. The simulated IAVs of aerosol concentrations over 2004−2012 in China were evaluated by Mu and Liao (2014). Our simulation by GEOS-Chem also captures the temporal variations of $PM_{2.5}$ concentrations in the eastern US, eastern China and Europe (see supplementary material, Fig. S2).

3. The meteorology-driven interannual variations of aerosols

We calculate the absolute percent departure from the mean (APDM) of $PM_{2.5}$ and its components to quantify the IAVs following Fu et al. (2012). The APDM is defined as:

$$\mathrm{APDM} = \frac{\frac{1}{n}\sum_{i=1}^{n}\left|C_i - \frac{1}{n}\sum_{i=1}^{n}C_i\right|}{\frac{1}{n}\sum_{i=1}^{n}C_i}\times 100\%,$$

where C_i is aerosol concentration in the i-th year, and n is the number of years examined. APDM represents the interannual variation relative to the average concentration over the n years. Figure 1 shows the geographic distribution of the APDM values of $PM_{2.5}$ concentrations in the eastern US, eastern China, and Europe. We will be focused on the APDM values in the polluted areas with the average $PM_{2.5}$ concentrations $\geq 5\ \mu g\ m^{-3}$. In the regions with low aerosol concentrations, the absolute variations of $PM_{2.5}$ concentrations are not large even though the APDM values are high.

As shown in Fig. 1, in the eastern US, the APDM values are 4−12% in most grid cells during DJF, MAM, JJA and SON. In eastern China, the APDM values exhibit a spatial pattern, with the maximum APDM values of 24−28% in DJF and of 32−36% in MAM in North China (NC). In other areas of eastern China, the APDM values are 4−12% during DJF and MAM, far less than

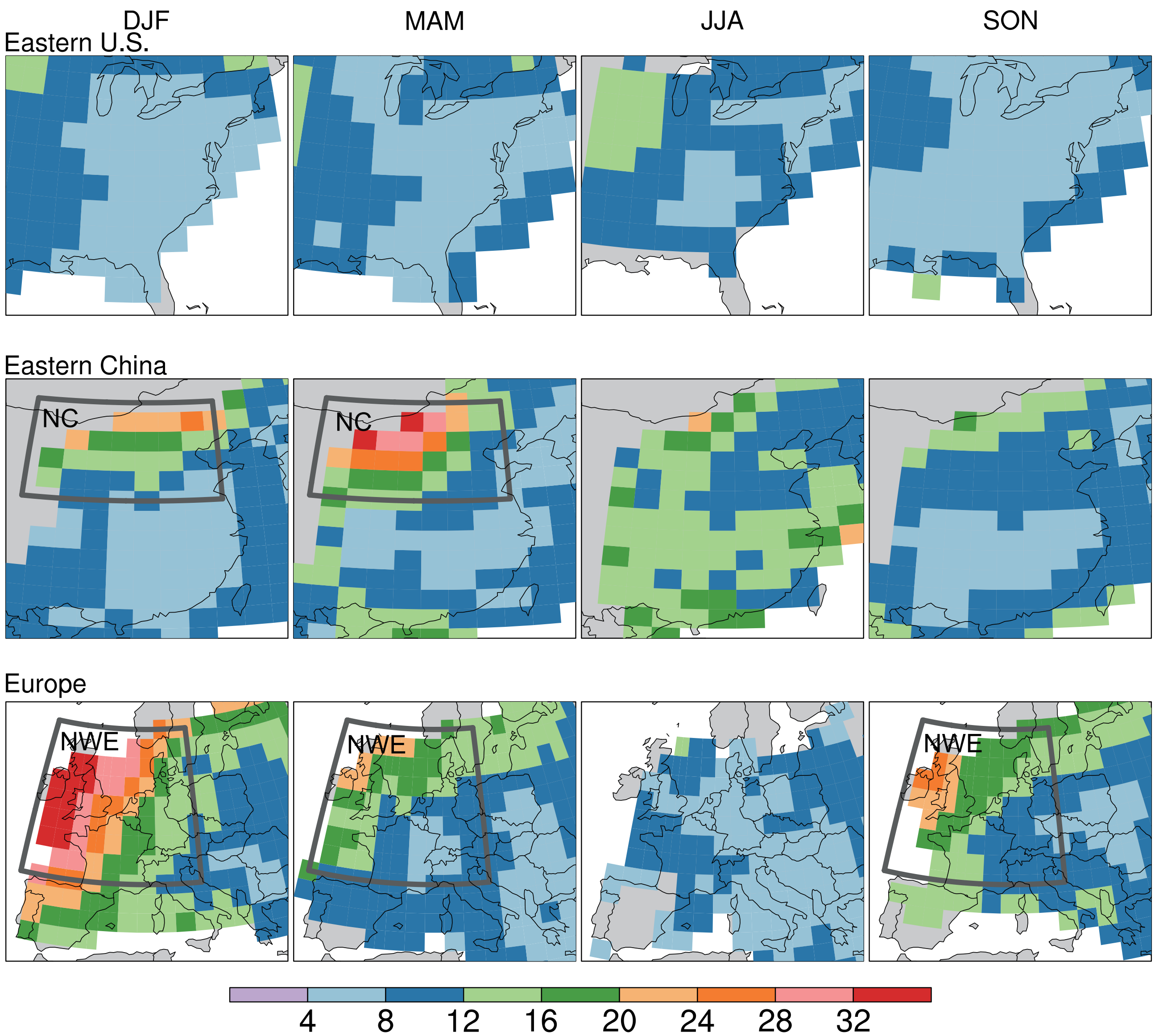

Fig. 1. The surface $PM_{2.5}$ APDMs (%) in the eastern US, the eastern China and Europe over DJF, MAM, JJA, and SON in 1986−2006. The grey polygons define North China (NC) and the northwestern Europe (NWE) respectively. Only the APDMs in the grids with the multiannual averaged concentration > 5 $\mu g\ m^{-3}$ are exhibited.

the values in NC. In JJA and SON, the IAVs in eastern China have more uniform geographic distributions, with APDM values of 8−20% in JJA and of 4−16% in SON in most grids. In JJA, the IAVs in eastern China are larger than in the eastern US and Europe. In Europe, the APDM values are high in the northwestern Europe (NWE) (in the Great Britain and Ireland), with the maximum values of 32−36%, 20−24% and 24−28% in DJF, MAM and SON, respectively. The APDM values are low (4−12%) in other areas of Europe.

In summary, we can draw conclusions that: (1) The IAVs of $PM_{2.5}$ in the eastern US are weaker and more uniform relative to those in eastern China and Europe. (2) In eastern China and Europe, the IAVs have obvious geographic patterns, high in the NC and NWE but low in other areas in DJF and MAM. (3) In JJA, the IAVs are quite uniform spatially in the eastern US, eastern China and Europe. We have calculated the APDM values of the observed $PM_{2.5}$ concentrations in the eastern US and Europe (Fig. S3 in supplementary material). Even though the IAVs of observed $PM_{2.5}$ concentrations are influenced by both local emissions and meteorological conditions, Fig. S3 shows that the IAVs of aerosols in the eastern US are uniform, similar to the simulated APDM values in Fig. 1.

4. Key meteorological variables that drive the interannual variations of $PM_{2.5}$ in the eastern US, eastern China and Europe

4.1 Methodology

We establish empirical relationship between main meteorological fields and aerosol concentrations by using the stepwise multi linear regression model (SLR) (Pratsinis 1988; Baek 1997; Lee et al. 2005; Yuan et al. 2006; Lin and Yeh 2007; Tai et al. 2010, see supplementary material). Among the 8 meteorological parameters of temperature at surface (TS), precipitation rate (PR), relative humidity (RH), surface wind speed (WS), air divergence at surface (DS), planetary boundary layer height (PBLH), and westerly wind at 850 hPa (U_{850}) and southerly wind at 850 hPa (V_{850}), the SLR identifies the statistically significant meteorological variables that have passed the F-test with 95% confidence. The WS, DS and PBLH are expected to modulate aerosol concentrations by vertical and horizontal ventilation. RH, PR and TS can impact aerosol concentrations by altering aerosol thermodynamics, wet deposition and chemical reaction rates (Dawson et al. 2007; Kleeman 2008; Jacob and Winner 2009). The wind fields at 850 hPa are considered here because the transport by the lower troposphere wind is important for surface-layer aerosol concentrations in the three studied regions (Zhang et al. 2010; Jerez et al. 2013; Gong et al. 2006).

We obtain the key meteorological parameters in the eastern US, eastern China and Europe by two steps: (1) In each grid cell, the SLR is used to get the time series of regressed $PM_{2.5}$ concentrations on the basis of the 8 meteorological variables and the statistically significant meteorological variables. (2) The APDM values are calculated for the regressed $PM_{2.5}$ concentrations (SLR APDM, see supplementary material). The geographic distributions of SLR APDM are then compared with those of the APDM values shown in Fig. 1.

4.2 Key meteorological variables that drive the interannual variations of $PM_{2.5}$ in the eastern US, eastern China and Europe

Figure 2 shows the SLR APDM values of $PM_{2.5}$ concentrations in the eastern US, eastern China and Europe. The geographic distributions and magnitudes of SLR APDM are similar to those of the APDM values shown in Fig. 2. Importantly, the SLR APDM values of $PM_{2.5}$ well reflect the three important features of APDM values mentioned in the end of Section 3. The similarity between the geographic distribution of APDM in Fig. 1 and that of the SLR APDM in Fig. 2 can be quantified by the pattern correlation coefficient (PCC, http://glossary.ametsoc.org/wiki/Pattern_correlation). The PCC in all four seasons and all three regions are always > 0.98 (Fig. 2), indicating that the IAVs of $PM_{2.5}$ reconstructed by SLR work well in all four seasons over the three regions.

Based on the SLR APDM, we also obtain the key meteorological variables in every grid cell by SLR. Then we define the meteorological variable importance index (MVII) for IAVs as:

$$\mathrm{MVII} = \frac{\text{the number of grids with a specific meteorological variable}}{\text{the number of all grids in the region}} \times 100\%$$

The MVII denotes the percentage of grid cells with a specific meteorological variable statistically significant in SLR. Figure 3 shows MVII in four seasons in the eastern US, eastern China, and Europe. Table S1 shows the most important meteorological variable (the meteorological parameter with the maximum MVII) obtained by SLR and its APDM value for a region (eastern US, eastern China, or Europe) and a season.

In the eastern US, the most important meteorological variable in DJF is U_{850}, with the MVII of 55%. Another two important variables are PBLH (MVII is 50%) and TS (MVII is 45%). In JJA, the most important meteorological variable is PR, with the MVII of 42%. In MAM and SON, the MVII values of PBLH are about 40% and 45% respectively, dominating the IAVs of $PM_{2.5}$ concentrations. The U_{850} drives the IAVs of $PM_{2.5}$ in DJF, because of the importance of wide-range transport due to the strong westerly in the lower troposphere (Feng et al. 2016) and the low photochemical activity during winter (Wagstrom and Pandis 2011a, b). The PR drives the IAVs of $PM_{2.5}$ in JJA, reflecting the importance of wet deposition during summer in the US. Dawson et al. (2007) reported that the sensitivities of $PM_{2.5}$ concentrations to precipitation were $-0.02\%\%^{-1}$ in January but $-0.2\%\%^{-1}$ in July in the eastern US because of the high PR in summer.

In eastern China, the most important meteorological variables in DJF and JJA are WS and RH, with the MVII values of 40% and 37%, respectively. The dominant effect of WS in winter over eastern China has been reported by many previous studies (Zhao et al. 2013; Mu and Liao 2014; Zhou et al. 2015; Qu et al. 2015; Li et al. 2015), as a result of the IAVs of the intensity of Eastern Asian Winter Monsoon (EAWM) (Niu et al. 2010; Zhou et al. 2015; Li et al. 2015). In summer, the intensive influences of RH on variations of aerosol concentrations have also been shown by many studies. Fu et al. (2014) presented that, for RH > 60%, haze events were more likely to happen during summertime in North China Plain. Using the process analyses, Mu and Liao (2014) demonstrated that humidity was a key meteorological parameter that influenced the IAVs of SO_4^{2-} in North China and NO_3^- in eastern China during JJA, by influencing the gas-phase formation and gas-to-aerosol partitioning, respectively.

In Europe, the most important meteorological variable in DJF (JJA) is PBLH (TS) with the MVII of 74% (41%). The effect of TS on the IAVs of $PM_{2.5}$ in summer results from the significant decrease of ammonium nitrate owing to the increase in temperature in Europe (Megaritis et al. 2013). In MAM and SON, the U_{850} and V_{850} are the most important variables with the MVII values of about 64% and 55%, respectively. Besides, U_{850} is also the second important variable in DJF (MVII is about 47%) and JJA (MVII is about 40%), indicating that the circulation in the free troposphere plays an important role in the IAVs of $PM_{2.5}$ (Fig. 3). Considering that the North Atlantic Oscillation (NAO) is the most important circulation pattern over Europe, the NAO might have contributed to the IAVs of surface aerosol concentrations in Europe. The impact of NAO on surface aerosol concentrations in Europe has been studied by Jerez et al. (2013) for years of 1970−1999.

5. Conclusion

Our study reveals the interannual variations (IAVs) of surface-layer $PM_{2.5}$ in the eastern US, eastern China, and Europe by using the GEOS-Chem simulation in years of 1986−2006. In the eastern US, IAVs are weak and uniform compared to those in eastern China and Europe. In eastern China, IAVs are high in North China in DJF and MAM. In Europe, IAVs are high in the

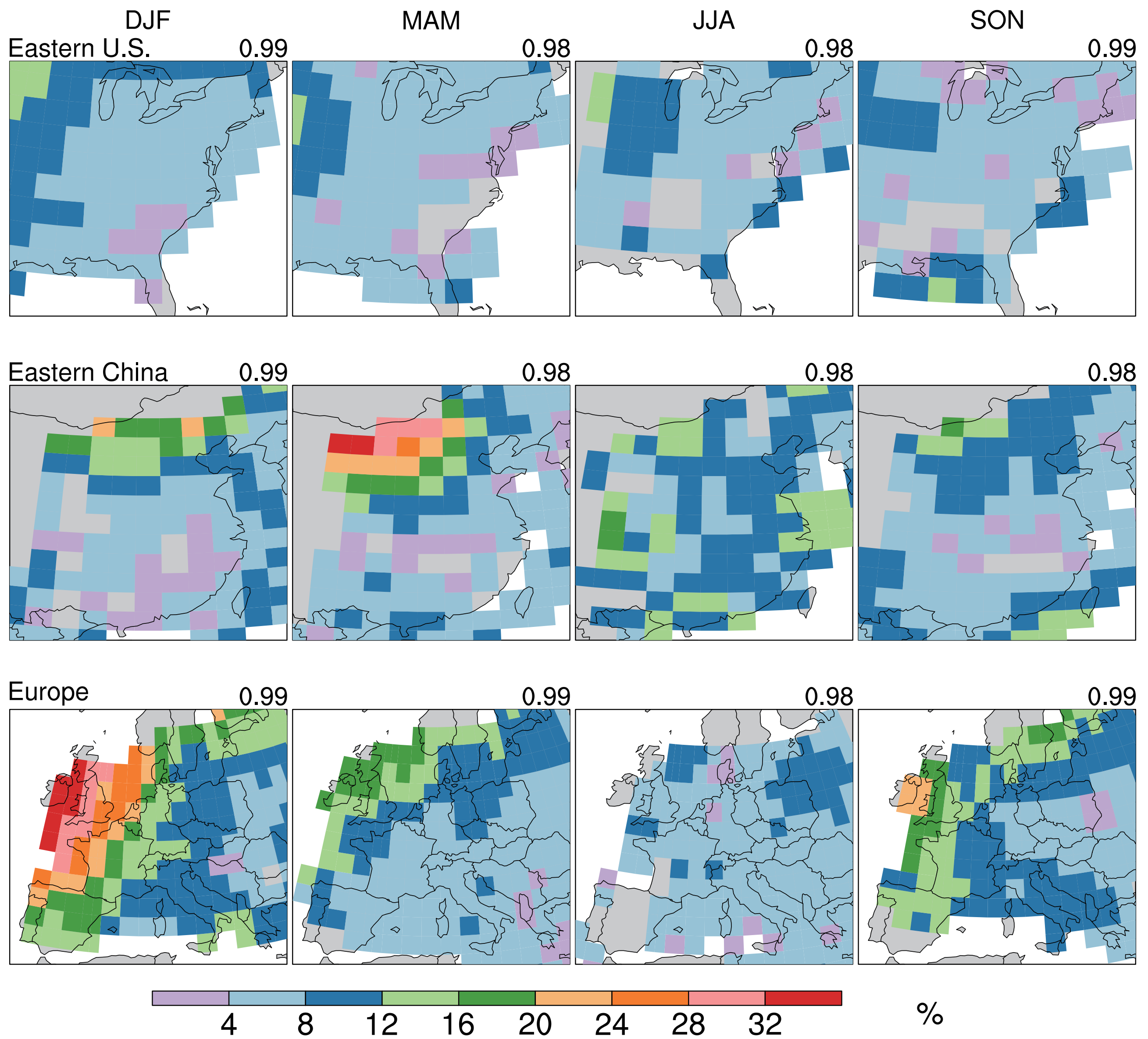

Fig. 2. Similar to Fig. 1 but for SLR APDM of $PM_{2.5}$. The numbers in the top right corner present the pattern correlation coefficients (PCC, see Section 4.2) between corresponding figures in Fig. 1 and Fig. 2.

northwestern Europe especially in DJF, MAM and SON. In JJA, the IAVs are uniform spatially in all the regions of the eastern US, eastern China and Europe. It should be note that the IAVs of aerosols reported in this study account for the role of variations in meteorological parameters alone. Mu and Liao (2014) reported that the variations in meteorology play more important roles than variations in anthropogenic emissions in driving the IAVs of aerosols over eastern China. Further studies are required to quantify the relative importance of variations in emissions and in meteorology in the IAVs of aerosols in the eastern US and Europe.

Using the stepwise multi linear regression (SLR) with 8 meteorological variables including temperature at surface (TS), precipitation rate (PR), relative humidity (RH), surface wind speed (WS), air divergence at surface (DS), planetary boundary layer height (PBLH), and westerly wind at 850 hPa (U_{850}) and southerly wind at 850 hPa (V_{850}), the key meteorological variables that drive the IAVs of $PM_{2.5}$ are obtained for each grid cell. The IAVs of $PM_{2.5}$ reconstructed by SLR agree with those of simulated aerosols in all four seasons over the three regions. By defining the meteorological variable importance index (MVII) based on SLR, we present the dominant meteorological variables for IAVs of $PM_{2.5}$ concentrations in the eastern US, eastern China, and Europe. In the eastern US, U_{850} and PR are the most important meteorological parameters in winter and summer, respectively. In eastern China, WS and RH are the most important meteorological parameters in winter and summer, respectively. In MAM and SON, the most important ones in China are RH and V_{850}, respectively. In Europe, PBLH and TS dominate in DJF and JJA, respectively. Additionally, U_{850} and V_{850}, which denote the atmospheric circulation in free troposphere, are always important variables in all the year round.

This study has some hints for assessment in the effectiveness of air quality control strategies in different regions and seasons. For example, since the eastern US has weaker and more uniform IAVs driven by meteorology, the IAVs in $PM_{2.5}$ concentrations can reflect well the effects of emission control strategies. On the contrary, in North China and the northwestern Europe in DJF and MAM, the effectiveness of air quality control strategies is possibly confused by the IAVs in $PM_{2.5}$ concentrations due to meteorology. Therefore, the IAVs in aerosol concentrations due to the dominant meteorological parameters should be concerned simultaneously in these regions and seasons in assessment of the effectiveness of air quality control strategies.

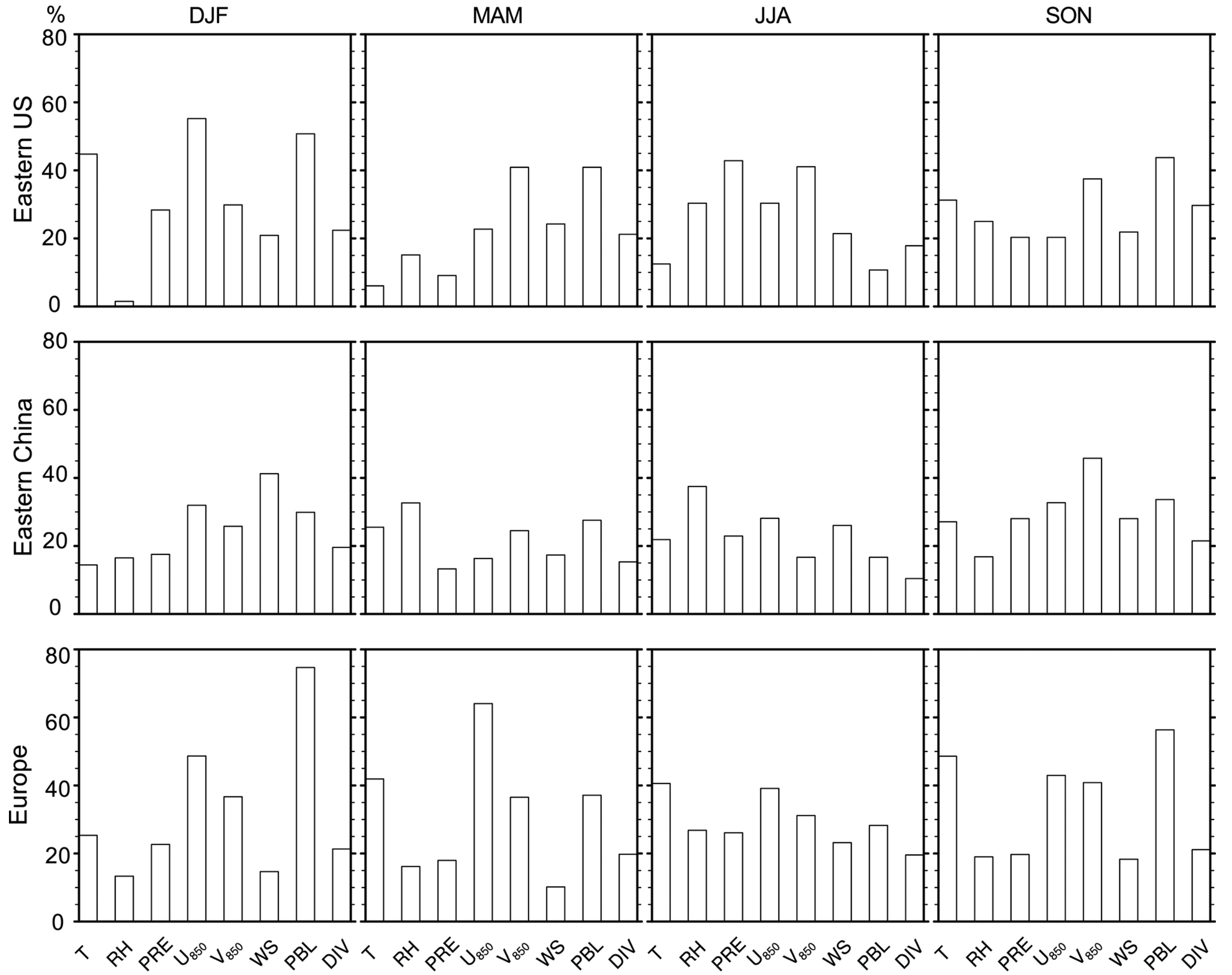

Fig. 3. The meteorological variable importance index (MVII, %, see Section 4.2) in the eastern US, the eastern China and Europe over DJF, MAM, JJA, SON in 1986−2006. The 8 meteorological parameters are temperature at surface (TS), precipitation rate (PR), relative humidity (RH), surface wind speed (WS), air divergence at surface (DS), planetary boundary layer height (PBLH), and westerly wind at 850 hPa (U_{850}) and southerly wind at 850 hPa (V_{850}).

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Supplement

Figures S1, S2, and S3, Table S1 and the description of the stepwise multi Linear regression (SLR) model and the SLR APDM are attached in the supplementary material.

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