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Spatial–temporal characteristics of haze and vertical distribution of aerosols over the Yangtze River Delta of China

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ABSTRACT

Variation of haze events occurred in the Yangtze River Delta (YRD) of China, the characteristics of meteorological elements and the vertical distribution of aerosols during haze episodes were analyzed by utilizing data of ground observation, radiosonde and CALIPSO. The results illustrate that the frequency of haze events between 1981 and 2010 peaked in winter but bottomed out in summer and decreased from north to south in the YRD region, reaching at the lowest point in "low frequency center" — Shanghai. When haze happened, the most seriously affected area was 2–4 km above the ground and the concentrated range of total backscattering coefficient (TBC) that decreased with altitude was 0.8×10^{-3}–2.5×10^{-3} $km^{-1}{\cdot}sr^{-1}$. Particulate depolarization ratio (PDR) was less than 40% in a large part and 93% aerosols over the YRD area were regular particles, while the irregular ones concentrated on 2 km above the surface and the irregularity rose up but the diversity diminished when altitude increased. Color ratio (CR) was lower than 1.2 mostly at all altitudes and distributed asymmetrically above the ground. Nearly 80% aerosols under 10 km were fine particles (CR < 1.0) and 22.54% coarse particles (CR > 1.0) clustered at 2–4 km. Large particles (CR > 1.2) aggregated in lower troposphere massively yet relatively smaller ones gathered in middle and upper troposphere. In the YRD region, aerosols with more powerful capabilities were wider and less regular than the ones of Northwestern China.

Introduction

Haze is one of air pollution phenomena, which is defined as visibility lower than 10 km when relative humidity is below 80% without precipitation, dust storm, blowing sand, floating dusts, smoke, blowing snow and snow storms (China Meteorological Administration, 2010) and whose higher frequency of occurrence in recent years and pernicious effects on human body have raised public concern as well as the scientific community in China (Zhang et al., 2012, 2015).

The Yangtze River Delta (YRD) region that covers about 99,600 km^2 with a population of 150 million, located in the east coast of China and including 16 core cities (the Shanghai municipality, 7 cities in the north of Zhejiang Province and 8 cities in the south of Jiangsu Province), contributed 18.5% of the national GDP in 2014. However, due to its rapid economic

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development with dramatic increased in energy consumption and pollutant emissions in the last 30 years, regional air pollution problems have become more and more salient, such as deteriorated urban air quality and declined visibility, all of which have made the YRD, together with the Pearl River Delta, Beijing–Tianjin–Tangshan and Sichuan–Chongqing, become one of the four heaviest haze regions in China (Fu et al., 2008; Wang et al., 2012; Cheng et al., 2013, 2014).

Previous studies have revealed that unfavorable meteorological elements, *e.g.*, high relative humidity, low rainfall, wind speed, atmospheric pressure, and extremely high concentrations of aerosol particles, including fine particulate matter, played critical roles in the formation of haze in the YRD area (Fu et al., 2008; Cheng et al., 2013; Wang et al., 2015a, 2015b; Zhang et al., 2015; Tian et al., 2016;). In addition, optical, microphysical properties, such as: size distribution, number concentration, and temporal–spatial distribution of aerosols during haze episodes were detected by ground-based field sampling measurements (Fu et al., 2008; Zhang et al., 2012, 2017; Cheng et al., 2013; Hu et al., 2014; Wang et al., 2014, 2015a, 2015b; Cui et al., 2016; Tian et al., 2016). Furthermore, numerical models were utilized to simulate the transport trajectory and forecast urban air quality in haze weather (Wang et al., 2012; Cheng et al., 2014), emission inventories of pollutants of the YRD region were also established (Huang et al., 2011; Fu et al., 2013). However, due to limitations of ground-based observations, data of vertical distribution of haze aerosols have still been difficult to achieve although it is an important part that cannot be neglected in estimating aerosol radiative forcing and its associated climate impacts (Claquin et al., 1998; Huang et al., 2008; Geng et al., 2011).

The CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) that was launched in 2006 can observe aerosols beneath thin clouds and provide aerosol profiles reflecting vertical structures over regional and global scales (Anselmo et al., 2005; Geng et al., 2011) while the data have mainly been exploited to analyze dust aerosols hitherto (Chen et al., 2010; Deng and Zhang, 2016; Wang et al., 2015a, 2015b, 2017) over northwestern China and researches on combining CALIPSO data with radiosonde data in the YRD area have still rarely been reported, which yet can endow us with two different opposite view angles on aerosols during haze periods when CALIPSO detects them straight down from sky to ground while radiosonde straight up from ground to sky.

Therefore, in this paper, we employed ground observations, satellite detection and aerological soundings to explore variation of haze events occurred over the YRD region, vertical structure of aerosol optical properties and meteorological factors in the boundary layer when haze formed, respectively. These analyses depict a new picture on haze and aerosols in the YRD so as to provide scientific basis for haze forecast.

1. Data and methods

1.1. Study domain

Seventeen meteorological stations (10 in Jiangsu Province, 2 in Shanghai municipality and 5 in Zhejiang Province) were selected to cover the area from 117.09°E to 121.27°E and 28.37°N to 34.17°N that represents the majority of the YRD area (Fig. 1a).

1.2. Ground observations and radiosonde data

All ground observations, including humidity and visibility, and radiosonde data, covering wind speed, direction, temperature and dew point temperature difference, from January 1981 to December 2016 of 18 meteorological stations (Fig. 1a), obtained from the China Meteorological Data Service Center and the Data Center of Ministry of Environmental Protection of China, were used to identify haze. Ground observations were operated four times a day at Beijing Time 02:00, 08:00, 14:00 and 20:00, respectively while radiosonde data were attained at Universal Time 00:00 and 12:00 only.

1.3. CALIPSO data

CALIPSO Level 2 Aerosol Profile Data Product both in daytime and nighttime condition from January 2014 to June 2016, including total backscatter coefficient (TBC) at 532 nm, particulate depolarization ratio (PDR) and backscatter coefficient at 1064 nm were utilized.

PDR is defined as the ratio of the perpendicular and parallel components of particulate backscatter coefficient at 532 nm, which displays the degree of irregularity of the detected particles; the higher the PDR is, the more irregular the particles are. Color ratio (CR) is defined as the ratio of backscatter coefficient at 1064 nm and that at 532 nm, reflecting the size of aerosols; the higher the CR is, the larger the particles are (Anselmo et al., 2005; Geng et al., 2011).

1.4. Methodology

After checking the data quality to ensure integrity and consistency, according to the statistical method of haze (China Meteorological Administration, 2010), we firstly summarized the 30-year spatial–temporal characteristics of haze in 12 stations (Fig. 1b) of the YRD between 1981 and 2010 and sifted 190 haze events from January 2014 to June 2016. Then the meteorological factors of 7 stations (Xuzhou, Sheyang, Nanjing, Baoshan, Hangzhou, Quzhou and Hongjia) and vertical distribution of aerosol optical properties during haze episodes were analyzed based upon the radiosonde data and CALIPSO, respectively.

2. Results and discussions

2.1. Spatial–temporal characteristics of haze

If there is no precipitation, dust storm, blowing sand, floating dust, smoke, blowing snow and snow storm on one certain day, meanwhile, visibility and relative humidity are below 10 km and 80%, respectively, then we determine it as one “haze day”. Variation of 30-year accumulated haze days (Fig. 1b) illustrates that the frequency of haze events between the period of 1981 and 2010 decreased generally from north to south over the YRD region although with an episodic steady growth in Zhejiang Province, reaching at the lowest point

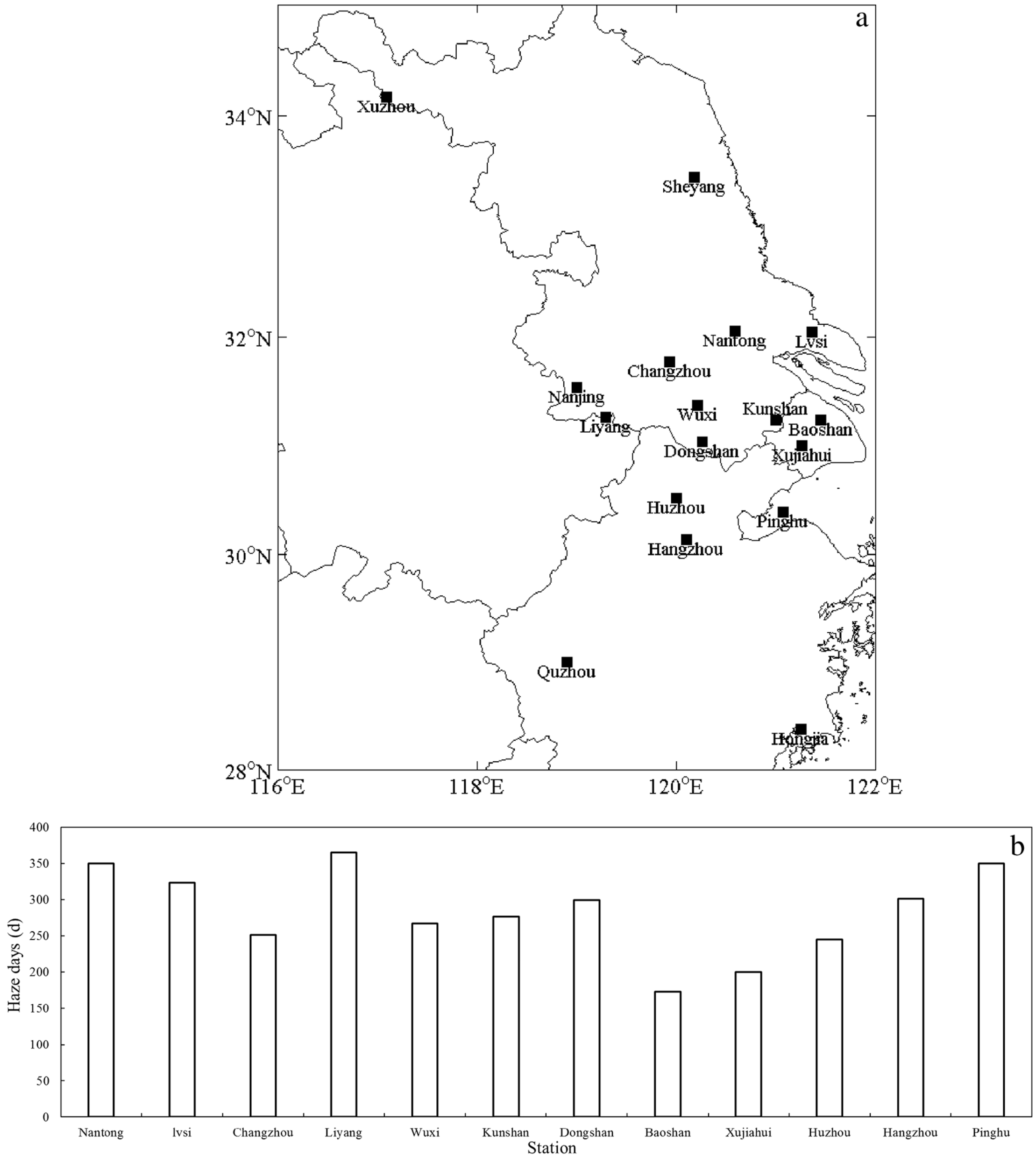

Fig. 1 – (a) Location of meteorological stations. Except Sheyang, Lvsi, Liyang, Kunshan, Dongshan and Hongjia are in the suburban area, all others lie in the urban center of each city. Baoshan and Xujiahui are located in Shanghai. (b) Variation of accumulated haze days at 12 stations over Yangtze River Delta from 1981 to 2010.

in Shanghai. These 30 years witnessed Shanghai as a "low frequency center", from which the haze frequency started to grow towards inland (Cheng et al., 2013).

Monthly variation of haze days in Jiangsu Province, Shanghai Municipality and Zhejiang Province (Fig. 2) all demonstrate that haze frequency tend to be highest in winter (November) but lowest in summertime (August), having a secondary peak in spring (April) and a secondary nadir in February.

In wintertime, massive amount of coal consumptions over the YRD area, especially from power plants, is the main reason for the highest haze days observed (Huang et al., 2011; Fu et al., 2013). In spring, sand–dust storm contributes a lot to tropospheric aerosol, which also leads to the formation of the secondary highest point of haze frequency (Zhang et al., 2012). Besides, the YRD is also the main agricultural production area across China, after wheat and food crop harvests, the scale of biomass burning, fertilizer and other agricultural activities definitely amplifies, which is the reason why the observed haze days show a general seasonal variation pattern with a summit in winter, falling down and then ascending in spring, bottoming out in summer and then rising up in autumn (Zhang et al., 2012; Cheng et al., 2014).

It is noteworthy that average haze days, derived from monthly variation, of Jiangsu Province, Shanghai Municipality and Zhejiang Province reflect the spatial characteristic of haze in the YRD as well. For instance, Shanghai has the minimum average haze days, corresponding to the "low frequency center" of the YRD region.

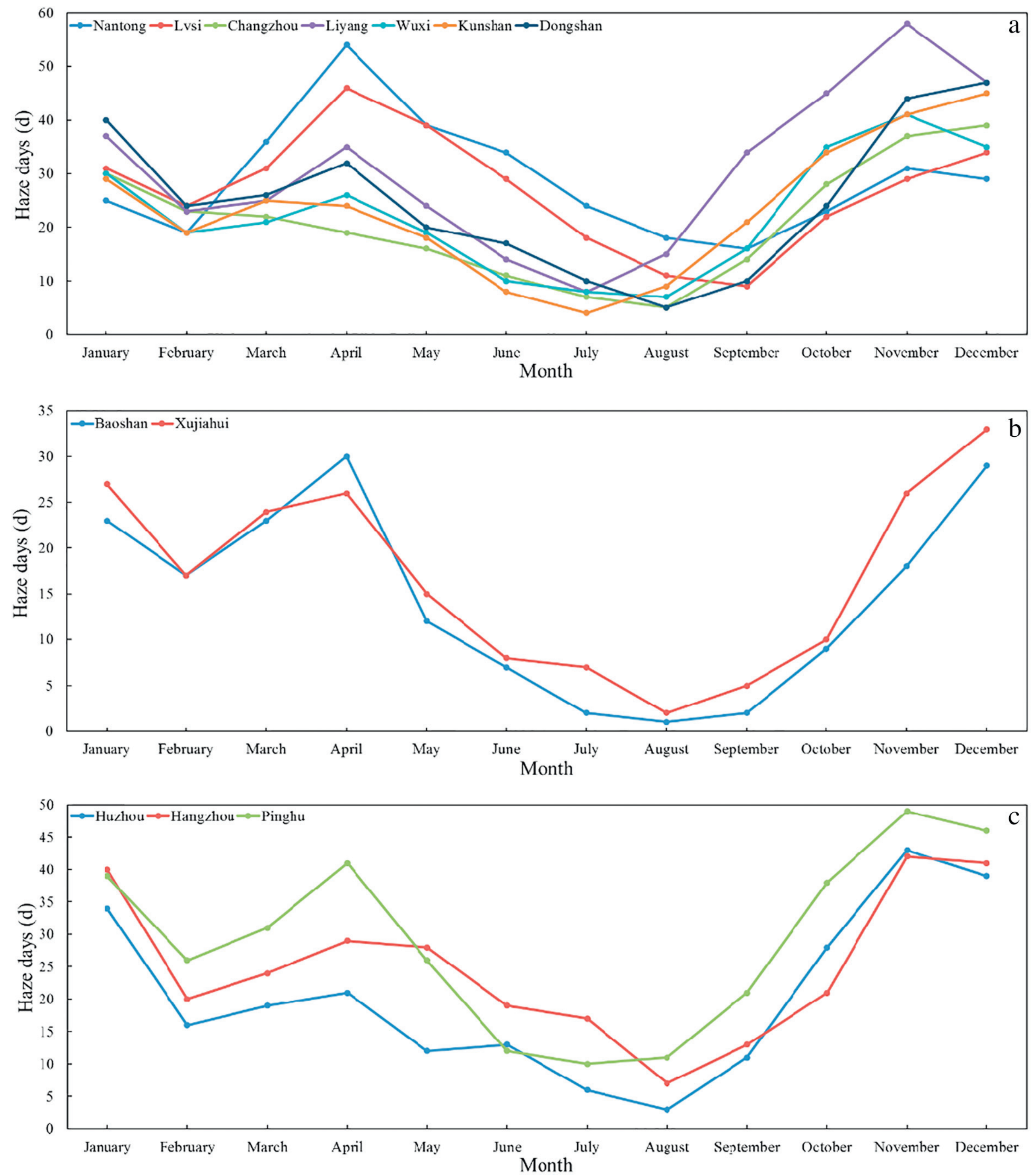

Fig. 2 – Monthly variation of haze days over (a) Jiangsu Province, (b) Shanghai Municipality and (c) Zhejiang Province from 1981 to 2010.

2.2. Characteristics of meteorological elements

2.2.1. Wind

Frequency distribution of wind direction (Appendix A Fig. S1) and speed (Appendix A Fig. S1) at different heights of boundary layer (1040–1000, 1000–900 and 900–850 hPa) were calculated. It is clear that wind direction of three layers at Universal Time 12:00 tends to be more stable than the ones at 00:00 over the YRD region except Baoshan and Hangzhou at which winds of two different directions (west *versus* south in Baoshan, east *versus* north in Hangzhou) share nearly the same occurrence rate because of strong turbulence between 1040 and 1000 hPa, resulting in the quick dissipation of haze. Besides, it could not be ignored that prevailing wind direction near the surface during haze episodes are around northwest in the northern YRD (Xuzhou and Sheyang) while east or east–northeast in the southern region (Nanjing, Baoshan, Hangzhou, Quzhou and Hongjia).

Wind speeds at Universal Time 00:00 and 12:00 share the same tendency that occurrence frequency of higher ones rises with altitudes and rates of lower ones (0–2 and 2–4 m/sec) remains around 0.6 at 1040–1000 hPa, under which circumstances, haze is more difficult to blow away.

No matter direction or speed, winds at 12:00 are unfavorable for dispersion of haze due to the weakening turbulence in boundary layer so that aerosol particles could accumulate massively and easily.

2.2.2. Temperature and humidity

Both temperature and humidity profiles (Appendix A Fig. S1) calculated by sounding data show evident inversions above surface and lower-level of boundary layer, which illustrate a stagnant dispersion atmosphere below 850 hPa. Due to convections and turbulence, temperature soundings and dew point temperature difference soundings at Universal Time 00:00 are more turbulent than ones at 12:00 during which a more stable atmosphere is formed. However, all of them expose a very strong surface inversion that trammels pollutants near the ground and a lower-level inversion that limits the vertical mixing of air is also detected from 1000 to 970 hPa. Moreover, at the same level, the air is comparatively constant humid near the ground since dew point temperature difference (3°C) is small, which promoted gas-to-particle transformations in the atmosphere (Fu et al., 2008).

2.3. Vertical distribution of aerosols

2.3.1. TBC

TBC is one of the main parameters of CALIPSO satellite reflecting the ability of aerosol scattering. It is widely acknowledged that the TBC at 532 nm ranging from 10^{-4}–8×10^{-4}, 8×10^{-4}–4.5×10^{-3} and 4.5×10^{-3}–10^{-2} $km^{-1} \cdot sr^{-1}$ represents gas molecules, aerosols and clouds, respectively (Liu et al., 2008).

Thus we picked up aerosol particulates and calculated the rate at each altitude of all samples (Fig. 3). It is obvious that most aerosols concentrate on the 2–4 km above the ground during haze periods, which accounts for 43.94%, followed by 30.3% aerosols floating under 2 km altitude, *i.e.*, the major polluted area in the haze events of YRD region is below 4 km height.

Table 1 reveals the vertical frequency distribution of TBC of aerosols. At the height of 0–2 km, aerosols with TBC ranging from 2.5×10^{-3}–4.5×10^{-3} $km^{-1} \cdot sr^{-1}$ account for 70% while at 2–4, 4–6 and 6–8 km that value mostly concentrate on the range of 0.8×10^{-3}–2.5×10^{-3} $km^{-1} \cdot sr^{-1}$ and from 8 to 10 km, aerosols' TBC only exists in the scope of 0.8×10^{-3}–1.5×10^{-3} $km^{-1} \cdot sr^{-1}$.

Table 1 – Percentage of aerosols with different total backscatter coefficient (TBC) at different altitudes.

TBC ($\times 10^{-4}$ $km^{-1} \cdot sr^{-1}$)	Altitude (km)				
	0–2	2–4	4–6	6–8	8–10
8–15	5.00%	37.93%	72.73%	75.00%	100.00%
15–25	25.00%	37.93%	27.27%	25.00%	0
25–35	30.00%	10.34%	0	0	0
35–45	40.00%	13.79%	0	0	0

With the rising of altitude, rate of aerosols with smaller TBC becomes larger while the one with bigger declines. Especially between 0 and 4 km above the ground, rate of aerosols with TBC ranging from 2.5×10^{-3}–4.5×10^{-3} $km^{-1} \cdot sr^{-1}$ falls down sharply from 70% to 24.13%. Hence, scattering of aerosols weakens from 0 to 10 km.

2.3.2. PDR

PDR which can distinguish spherical aerosols from non-spherical aerosols is another data product of CALIPSO. As illustrated by Table 2, almost 85% aerosols' PDR value ranges from 0% to 40% at all altitudes; exactly, regular aerosols with the value of PDR less than 60% account for 93% from 0 to 10 km while irregular ones with larger values (PDR > 60%) cluster beneath 2 km above the ground and distribute at all levels except 8–10 km.

When the altitude increases, rate of aerosols with lower PDR descends while those with higher PDR ascends on the whole, which means that aerosols are more regular in lower troposphere than those in middle and upper troposphere when haze episodes occur in the YRD area. In other words, the irregularity of aerosols aggrandizes with altitudes as a whole, on the contrary, diversity of aerosols with different PDR diminishes due to impact of power-plant plumes, local emissions and burst of nucleation processes near the surface, which could enrich aerosol concentrations and types (Gao et al., 2009).

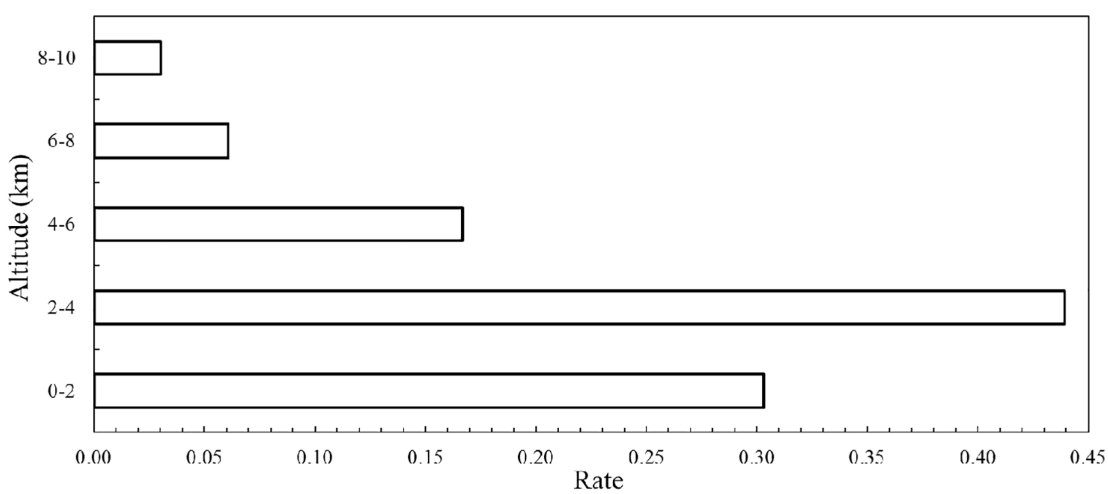

Fig. 3 – The rate distribution of aerosols at different heights during haze periods.

Table 2 – Percentage of aerosols with different particulate depolarization ratio (PDR) at different altitudes.

PDR (%)	Altitude (km)				
	0–2	2–4	4–6	6–8	8–10
0–5	8.62%	4.08%	3.85%	22.22%	10.00%
5–10	20.69%	22.45%	15.38%	11.11%	0
10–15	25.86%	18.37%	30.77%	11.11%	10.00%
15–20	5.17%	18.37%	7.69%	5.56%	10.00%
20–30	8.62%	16.33%	19.23%	11.11%	30.00%
30–40	5.17%	10.20%	11.54%	22.22%	30.00%
40–50	6.90%	4.08%	0	0	10.00%
50–60	3.45%	2.04%	7.69%	5.56%	0
60–70	8.62%	0	0	0	0
70–80	1.72%	4.08%	3.85%	5.56%	0
80–90	3.45%	0	0	0	0
90–100	1.72%	0	0	5.56%	0

According to the analysis of dust distributions in Tibet Plateau (Liu et al., 2008), PDR of dust aerosols ranges from 10% to 35% while that of maritime aerosols is near zero because most of them are spherical ones, and maximum PDR of smoke aerosols including black carbon aerosols which are close to spherical is about 2%. Therefore, based upon Table 2, it can be determined that dust, smoke and maritime aerosols are the major types in lower troposphere; the proportions of spherical and non-spherical aerosols are nearly the same in lower and middle troposphere; due to the dusts transported from northwest, local convections and building construction, aerosols whose PDR is about 5%–15% appear equally in upper troposphere (Fu et al., 2008; Liu et al., 2012; Wang et al., 2015a, 2015b).

2.3.3. CR

As reflected by Fig. 4, almost CR of aerosols at all altitudes is less than 1.2 when haze happens although each interval distributes asymmetrically. Fine particles whose CR is inferior to 1.0 account for 80% approximately within 10 km height while rate of coarse particles with CR higher than 1.0 increases slightly, specifically at 2–4 km with the maximum 22.54%. Proportion of aerosols whose CR changes from 1.0 to 1.2 goes up with the ascent of altitude continuously till the top value at 8–10 km where aerosols with 0.6–0.8 CR have the greatest ratio (60%) as well, on the contrary, that from 0.4 to 0.6 is the largest beneath 2 km off the ground. Generally, due to gravity action, large particles whose CR is higher than 1.2 mostly concentrate in lower troposphere, *e.g.*, 11.27% aerosols' CR between 2 and 4 km is centered around 1.4; in middle and upper troposphere, however, relatively small particles aggregate massively, which demonstrate the overall trend that aerosols' particle size drops down with the rising of altitudes.

It is well known that CR of ordinary aerosols varies from 0 to 0.5 that are smaller than the one of maritime aerosols which concentrates on 0–0.75 and CR of smoke aerosols distributes from 0 to 1.5 while dust aerosols are the largest one among all types, whose CR ranges from 0.25 to 1.25 because of the largest size of dust particles (Liu et al., 2008).

Therefore it can be summarized that smoke and dust aerosols exist widely in lower troposphere while ordinary and maritime aerosols dominate in middle and upper troposphere. Below 10 km above the surface in the YRD region, particularly at 0–2 and 8–10 km, aerosols with CR from 0.4–0.8 occupy dominant position during haze periods and they are extremely probably dust aerosols whose CR peaks at 0.8.

Table 3 – Comparison of TBC, PDR and CR over the northwestern China and Yangtze River Delta when haze occurred.

	TBC	PDR	CR
Northwestern China	0.8×10^{-3}–2.5×10^{-3} km^{-1}·sr^{-1}	0%–20%	0.0–0.8
Yangtze River Delta	0.8×10^{-3}–3.0×10^{-3} km^{-1}·sr^{-1}	0%–40%	0.0–1.2

TBC: total backscatter coefficient; PRD: particulate depolarization ratio; CR: color ratio.

2.4. Comparison

Few studies (Liu et al., 2008; Yan et al., 2016) have disclosed vertical distribution of aerosols (TBC, PDR and CR) over the northwestern China, but we still could compare results with them to highlight characteristics of aerosols in the YRD.

As shown by Table 3, aerosols are all with higher TBC, PDR and CR in the YRD region than the ones of northwestern China,

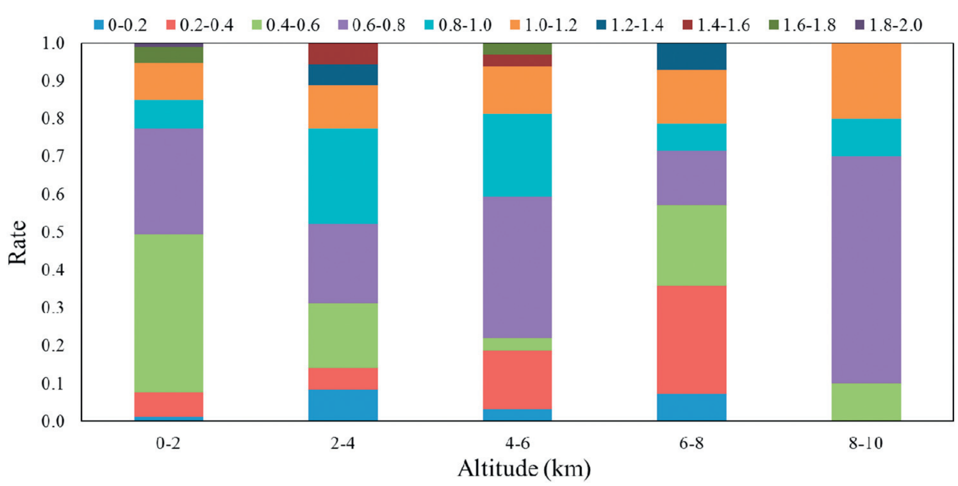

Fig. 4 – Histogram of frequency distribution of color ratio at different altitudes during haze periods.

which means that more irregular and larger aerosol particles, with stronger scattering abilities, exist in the YRD.

Compared with less population density and lower economic growth in the Northwestern China, the YRD, as a heavy industries cluster and large agricultural production area, has higher aerosol emissions from automobile manufacturing, power plants and biofuel burning, *etc.*, which is superimposed by unusual atmospheric circulation, depression of cold air activities and stagnant dispersion, leading to more complicated categories of aerosol particles and subsequent particle growth in haze episodes, resulting in greater values of TBC, PDR and CR (Fu et al., 2008; Gao et al., 2009; Zhang et al., 2012; Cheng et al., 2014). In fact, aerosols derive from industrial sectors, mobile sources, dust and sea salt in the YRD region while the northwestern ones mainly from residential and industrial sectors (Zhang et al., 2012; Wang et al., 2014; Yan et al., 2016).

3. Conclusions

From 1981 to 2010, haze occurrence rate in the YRD that tended to be highest in winter (November) but lowest in summertime (August) while having a secondary summit in spring (April) and a secondary nadir in February decreased generally from north to south until reaching the "low frequency center" — Shanghai where the haze frequency started to grow towards inland. Massive amount of coal consumptions over the YRD area in wintertime, especially from power plants, sand–dust storm that contributes a lot to tropospheric aerosol in spring and as a main agricultural production area across China, definitely amplified scale of biomass burning, fertilizer and other agricultural activities after wheat and food crop harvests, all of them are the reasons why the observed haze days follow a general seasonal variation pattern with a peak in winter, dropping down and then ascending in spring, bottoming out in summer and then rising up in autumn.

When haze happens, due to the weakening turbulence, a stable and stagnant dispersion atmosphere that is unfavorable for dissipation of haze exists within the boundary layer (1030–850 hPa): northwestern winds tend to be prevailed with a speed of 2–4 m/sec at Universal Time 12:00 p.m.; between 1000 and 970 hPa, evident inversions trammel pollutants near the ground and limit the vertical mixing of air, where the humidity is comparatively constantly high, which promoted gas-to-particle transformations in the atmosphere. Under that circumstances, aerosol particles accumulate massively and easily.

During haze periods in the YRD region, 74.24% aerosols concentrate below 4 km height within which aerosols with TBC ranging from 2.5×10^{-3}–4.5×10^{-3} $km^{-1} \cdot sr^{-1}$ account for 70% at the height of 0–2 km, which makes there to be the major polluted area. Aerosols' TBC that concentrates between 0.8×10^{-3} and 2.5×10^{-3} $km^{-1} \cdot sr^{-1}$ falling down with altitude discloses that scattering of aerosols weakens from 0 to 10 km in the vertical direction.

In addition, almost 85% aerosols' PDR is centered around 0%–40% at all altitudes and regular aerosols account for 93% from 0 to 10 km while irregular aerosols cluster beneath 2 km above the ground and distribute at all levels apart from 8–10 km. Irregularity of aerosols aggrandizing with altitudes as a whole is reflected by that rate of aerosols with lower PDR descends while those with higher one ascends with the rising of altitude when haze events occur over the YRD area.

Moreover, CR is lower than 1.2 mostly at all altitudes although each interval distributes asymmetrically above the ground. Fine particles account for nearly 80% within 10 km height and 22.54% coarse particles cluster at 2–4 km. Generally, large particles aggregate mostly in lower troposphere; yet in middle and upper troposphere, relatively small particles gather massively, in other words, aerosols' particle size drops down when varied with altitudes.

When haze occurs in the YRD, dust and smoke aerosols occupy dominant position in lower troposphere and maritime aerosols are the major type in middle and upper troposphere, spherical and non-spherical aerosols share nearly the same ratios in lower and middle troposphere. In comparison with Northwestern China, aerosol particles in the YRD have stronger scattering ability, higher irregularity and larger size due to various particle categories and particle growth.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2017.05.039.

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