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# Contrasting trends of mass and optical properties of aerosols over the Northern Hemisphere from 1992 to 2011

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## Abstract

Atmospheric aerosols impact both human health and climate. PMX is the mass concentration of aerosol particles that have aerodynamic diameters less than X µm, $PM_{10}$ was initially selected to measure the environmental impact of aerosols. Recently, it was realized that fine particles are more hazardous than larger ones and should be measured. Consequently, observational data for $PM_{2.5}$ have been obtained but only for a much shorter period than that of $PM_{10.}$ Optical extinction of aerosols, the inverse of meteorological visibility, is sensitive to particles less than 1.0 µm. These fine particles only account for a small part of total mass of aerosols although they are very efficient in light extinction. Comparisons are made between $PM_{10}$ and $PM_{2.5}$ over the period when the latter is available and with visibility data for a longer period. $PM_{10}$ has decreased by 44 % in Europe from 1992 to 2009, 33 % in the US from 1993 to 2010, 10 % in Canada from 1994 to 2009, and 26 % in China from 2000 to 2010. However, in contrast, aerosol optical extinction increased 7 % in the US, 10 % in Canada, and 18 % in China during the above study periods. The reduction of optical extinction over Europe of 5 % is also much less than the 44 % reduction in $PM_{10.}$ Over its short period of record $PM_{2.5}$ decreased less than $PM_{10.}$ Hence, $PM_{10}$ is neither a good measure of changes in smaller particles or of their long-term trends, a result that has important implications for both climate impact and human health effects. The increased fraction of anthropogenic aerosol emission, such as vehicle exhaust, to total atmospheric aerosols partly explains this contrasting trend of optical and mass properties of aerosols.

## 1 Introduction

Atmospheric aerosols are a mixture of solid and aqueous species that enter the atmosphere by anthropogenic and natural pathways (Querol et al., 2004). They can be directly emitted from power plants, motor vehicles, industrial facilities, and natural sources, such as dust and ocean foam (Andreae et al., 2009). They can also be formed





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photo-chemically from reactions of primary gaseous species in the atmosphere, including $SO_2$ and $NO_2$ (Streets et al., 2007). Aerosols scatter and absorb solar radiation, reducing surface incident solar radiation and heating the aerosol layer (Ramanathan et al., 2007), hence changing atmospheric circulation and the water cycle (Rosenfeld et al., 2008; Kaufman et al., 2002).

$PM_{10}$ has been selected by various Environmental Protection Agencies (EPAs) to measure environmental impact. The World Health Organization (WHO) recently announced that fine particles are found to be more hazardous than larger ones in terms of mortality and cardiovascular and respiratory endpoints in panel studies (Englert, 2004). The loading of such fine particles could be estimated by measurement of $PM_{2.5}$ or $PM_{1.0}$ (Watson, 2002; Nicole Pauly, 2009). The impact of atmospheric aerosols on visibility through their scattering and absorption of solar radiation is especially sensitive to fine particles, i.e., those less than 1.0 µm (Watson, 2002; Nicole Pauly, 2009), because these particles, comparable in size to the wavelength of visible solar radiation have the largest optical extinction efficiency. The inverse of optical extinction, meteorological visibility is routinely observed at meteorological stations and is available from more than 9000 such sites. Visibility has been used successfully to quantify long-term variation of aerosols during the past four decades (Field et al., 2009; Vautard et al., 2009; Wang et al., 2009). In summary, both health and climate are most strongly dependent on the fine particles with sizes of a few microns or less.

Because the longest records and greatest numbers of observational sites are for the larger particles a key question is whether they could provide an adequate measure of the smaller particles, and hence health and climate effects. This question can be addressed using $PM_{10}$ measurement collected in Europe, the US, Canada, and China from 1992 to 2010 and comparing them with optical extinction measurements, and with the more limited $PM_{2.5}$ observations available since 1997. When comparing the optical and mass concentration of aerosols, normalized values are used to account for their different definition.





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## 2 Study regions and data

In this study, we investigate the long-term variation of optical and mass properties in the Northern Hemisphere, including China, the US, Canada, and Europe. We use $PM_{10}$ and $PM_{2.5}$ data to quantify long-term variation of mass property of atmospheric aerosols. The meteorological visibility is used to characterize the climatic variability of their optical extinction.

### 2.1 Visibility and meteorological data

This work uses the Global Summary of Day (GSOD) database distributed by the National Climatic Data Center (NCDC), USA (ftp://ftp.ncdc.noaa.gov/pub/data/gsod). Data from over 9,000 stations are typically available. The GSOD data contain 18 surface meteorological parameters derived from synoptic hourly observations: mean temperature, mean dew point, mean sea-level pressure, mean station pressure, daily mean visibility, mean wind speed, maximum sustained wind speed, maximum wind gust, maximum temperature, minimum temperature, precipitation amount, and snow depth. Information is also included on the occurrence of fog, rain/drizzle, snow/ice pellets, hail, thunder, and tornado/funnel clouds.

Visibility, the maximum distance at which an observer can discern the outline of an object against the horizon sky, is reduced mainly by the presence of aerosols and hydrometeors. Eliminating the influence of hydrometeors on visibility allows an estimation of the near-surface optical extinction coefficient of aerosols from long-term observations of visibility (Husar et al., 2000). In this study, we also corrected the impact of relative humidity on visibility according to the methods reported by Rosenfeld et al. (2007) and Che et al. (2007).

Meteorological visibility is observed by manual assessment in China, Europe, and Canada. This was also the case for the US before the 1990s. During the 1990s, the US used instrumental (visibility meter) observations to replace the manual assessment of visibility. These observations are not homogeneous and therefore, we do not use





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the manual assessment of visibility but retained the instrument observations to provide detailed information on the relationship between visibility and the optical extinction coefficient of aerosols.

In this study, we use the inverse of the visibility as a proxy of optical extinction of atmospheric aerosols. It has been successfully used to characterize long-term variation of atmospheric aerosols globally over the land from 1973 to 2007 (Wang et al., 2009). The long-term variability of atmospheric aerosols derived from visibility has been confirmed by numerous independent estimates of atmospheric aerosols using direct measurements (Mitchell et al., 2010; Yoon et al., 2012), satellite retrievals (Lau and Kim, 2010; Dey and Di Girolamo, 2011), radiation measurements (den Outer et al., 2010; Ruckstuhl et al., 2010; Dwyer et al., 2010), and global chemical model simulations (Pozzoli et al., 2011; Chiacchio et al., 2011). Visibility has also been used to quantify long-term variation of haze over Europe (Vautard et al., 2009) and forest fire smoke over Southeast Asia (Field et al., 2009). The values of inverse of the visibility are averaged into monthly values when comparing with $PM_{10}$ and $PM_{2.5}$ data. Before the comparisons, we also normalized both mass concentration and optical extinction by their multi-year averages.

## 2.2 $PM_{10}$ data

### 2.2.1 $PM_{10}$ data over the US

For the US, hourly $PM_{10}$ measured by the Air Quality System from 1993 to 2010 was obtianed via the US Environmental Protection Agency website at: http://www.epa.gov/ttn/airs/airsaqs/. Data collected from approximately 800 sites were available. In this study, daily $PM_{10}$ was calculated from the hourly data, from which the monthly values were calculated. The monthly $PM_{10}$ was regarded as reliable only if the daily $PM_{10}$ values were available for more than 15 days during a month. We selected 129 sites where reliable monthly $PM_{10}$ data were available for more than 120 months from 1993 to 2010. Among the 129 sites, 51 sites were located in urban areas, 60 sites were





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located in suburban areas, and 18 sites were located in rural areas. See Figs. 1 and 2 for their locations and their site-average $PM_{10}$ values.

### 2.2.2 $PM_{10}$ data over Europe

Hourly $PM_{10}$ data over Europe were downloaded from AirBase: http://acm.eionet.europa.eu/databases/airbase/. About 3000 sites in the 39 European countries supplied $PM_{10}$ data from 1992 to 2009. Among them, 250 sites have more than 120 months of reliable monthly $PM_{10}$ data, including 148 urban sites, 62 suburban sites, and 40 rural sites. Figures 1 and 2 show their locations and their site-average $PM_{10}$ values.

### 2.2.3 $PM_{10}$ data over China

Daily $PM_{10}$ data over China were calculated from the daily Air Pollution Index (API) (from noon to noon), which were released by the Data Center of Ministry of Environmental Protection of the People's Republic of China: http://datacenter.mep.gov.cn/ (in Chinese). Hourly $PM_{10}$ data were collected at several sites in each city, for example, 8 sites for Beijing (Zhu et al., 2011). Data for $PM_{10}$, $NO_2$, and $SO_2$ were averaged to daily values and normalized to an API for each constituent. The highest of these API values were used to estimate the city API and released to the public.

The daily $PM_{10}$ values were estimated from API equations, assuming $PM_{10}$ was the primary pollutant and may be overestimated if the API was determined from $SO_2$ or $NO_2$ concentrations. We evaluated this uncertainty at 31 major cities in China, including province capitals and four municipalities. Annual averaged $PM_{10}$ values from 2003 to 2009 at the 31 mega-cities are available from the annual report of the National Bureau of Statistics of China (available at http://www.stats.gov.cn/tjsj/ndsj/, in Chinese). We calculated their city-average $PM_{10}$ values and compared them to those calculated from the API. The results are shown in Fig. 3. The overestimation (bias) is less than 2 %, with $R^2 = 0.995$. $PM_{10}$ is the primary pollutant for more than 80 % of the conditions in most cities. The cities in China range from those most polluted to those least polluted





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(Figs. 2 and 3). Therefore, we believe our $PM_{10}$ values calculated from the API are reliable. There are some cities where the API calculated $PM_{10}$ values are a little less than those averaged from the measurements. This is because when the calculated API is larger than 500, the API is set to 500 and that value is reported to the public. However, the measurements are only released at annual time scales for the 31 major cities. We have to use API to calculate PM10 in China at daily and monthly time scales.

API data are available at http://datacenter.mep.gov.cn/ for approximately 120 Chinese cities from 2000 to 2011. Eighty-six cities were selected where reliable monthly $PM_{10}$ data are available for more than 60 months from 2000 to 2011 (See Fig. 2), and half of them have $PM_{10}$ data for more than 120 months.

#### 2.2.4 $PM_{10}$ data over Canada

Hourly $PM_{10}$ data in Canada were collected through the National Air Pollution Surveillance (NAPS) Network: http://www.etc-cte.ec.gc.ca/napsdata/Default.aspx. The NAPS Network was established in 1969 as a joint program of the federal and provincial governments to monitor and assess ambient air in Canadian urban centres. The goal of the NAPS program is to provide accurate and long-term air quality data of a uniform standard throughout Canada. These $PM_{10}$ data are available from 1992 to 2009 at the above website. There were 110 sites that reported $PM_{10}$ to NAPS, and about 64 sites have more than 60 months of reliable monthly $PM_{10}$ data.

NAPS classified its stations into six types, Agricultural Rural (A), Commercial (C), Forested Rural (F), Industrial (I), Residential (R), and Undeveloped Rural (U). For consistency with other regions, we reorganized them into three basic types (Fig. 1): C and R are urban types, A, F and U are rural stations, and I is the suburban type. Most of the Canadian stations are located in urban areas. The detailed site information can be found in Figs. 1 and 2.





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### 2.3 $PM_{2.5}$ data

$PM_{2.5}$ data were available for the US, Canada, and European countries. China does not have routine $PM_{2.5}$ observations.

#### 2.3.1 $PM_{2.5}$ data over US

The US hourly $PM_{2.5}$ data are available from http://www.epa.gov/ttn/airs/airsaqs/. Approximately 900 stations reported $PM_{2.5}$ data from 1993 to 2010. A total of 406 stations were selected where reliable monthly $PM_{2.5}$ data were available for more than 60 months from 1998 to 2010, including 160 urban stations, 160 suburban stations, and 68 rural stations. See Figs. 1 and 2 for detailed information.

#### 2.3.2 $PM_{2.5}$ data over Europe

Hourly $PM_{2.5}$ data for Europe from 1998 to 2009 are available at http://acm.eionet.europa.eu/databases/airbase/. About 700 stations reported $PM_{2.5}$ data. In this study, 98 stations were selected where reliable monthly $PM_{2.5}$ data were available for more than 60 months from 1998 to 2009, including 53 urban stations, 17 suburban stations, and 28 rural stations. Figures 1 and 2 provide detailed information.

#### 2.3.3 $PM_{2.5}$ data over Canada

Hourly $PM_{2.5}$ data in Canada were collected through the NAPS Network: http://www.etc-cte.ec.gc.ca/napsdata/Default.aspx. These $PM_{2.5}$ data are available from 1995 to 2009. A total of 159 sites reported $PM_{10}$ to NAPS, and about 56 sites have more than 60 months of reliable $PM_{2.5}$ data. Most of the sites are located in urban areas. The site information can be found in Figs. 1 and 2.





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### 2.4 $PM_{10}$ and $PM_{2.5}$ measurement methods and data consistency

Reference methods for $PM_{10}$ and $PM_{2.5}$ mass concentrations includes the European reference method (EN 12341 NORM) and the US EPA reference method for $PM_{10}$ (http://www.epa.gov/ttn/emc/promgate/m-201.pdf). Alternative methods for $PM_{10}$ or $PM_{2.5}$ may introduce positive and negative sampling artifacts comparing with the reference methods, depending on chemical composition of the aerosols, as well as meteorological conditions (Putaud et al., 2004, 2010).

One of the primary factors resulting in data inconsistency is the relative humidity (RH) requirement of different measurement methods. For example, at a continental site (Putaud et al., 2004), $PM_{10}$ and $PM_{2.5}$ measurements at 50 % RH may be 9 % and 7 % higher than those at 20 % RH. Studies show that the routine tapered element oscillating microbalancemethod underestimates $PM_{10}$ measurements by up to 35 %, when compared with the European reference method (Allen et al., 1997; Van Dingenen et al., 2004). This underestimation is more severe in winter than in summer.

To keep data consistent, we selected data from one measurement method for each European, US, and Canada site. However, we do not have information on the method of measurement of $PM_{10}$ used at the Chinese sites. In this paper, we focus on the long-term variation of $PM_{10}$ and $PM_{2.5}$ and normalized values are discussed below.

## 3 Results

### 3.1 Climatology and long-term trends of $PM_{10}$

As for multi-year average of $PM_{10}$, China's urban areas are the most polluted, while Canada is the least polluted region studied (Table 1 and Fig. 2). Table 1 also demonstrates that there are no substantial differences in the $PM_{10}$ concentration over urban, suburban, and rural areas in Europe and the US, most likely because urban areas in European countries and the US are lightly polluted in terms of $PM_{10}$.





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$PM_{10}$ substantially decreased in Europe, the US, and Canada from 1992 to 2011 (Fig. 4). $PM_{10}$ in Europe decreased by 13 $\mu g\,m^{-3}$ (44 %), averaged from 250 sites from 1992 to 2009, with peaks in 1996, 2003, and 2006. The 2003 European summer heat wave caused the record-breaking forest fires in Portugal. Smoke from the forest fires (Garcia-Herrera et al., 2010; Trigo et al., 2006) contributed to the $PM_{10}$ and $PM_{2.5}$ in 2003. However, these peak values were not reproduced in recent modeling studies, although most models predicted a long-term reduction of $PM_{10}$ similar to that observed during the study period (Chiacchio et al., 2010). The model did not include the forest fires (Chiacchio et al., 2010).

$PM_{10}$ in the US decreased by about 9 $\mu g\,m^{-3}$ (33 %), averaged over 129 sites from 1993 to 2010, with no large differences in the long-term trends in the urban, suburban, and rural areas of Europe and the US (Fig. 4), implying that emission controls worked equally well in all areas (Murphy et al., 2011). It decreased in Canada by about 2.3 $\mu g\,m^{-3}$ (15 %), averaged over 64 sites from 1994 to 2009. The more recent reduction in China, by 32 $\mu g\,m^{-3}$ (26 %), is attributed to emission controls, as energy consumption has increased dramatically since 2000 (Fig. 5).

### 3.2 Climatology and long-term trends of $PM_{2.5}$

$PM_{2.5}$ data are available from about 550 sites in Europe, the US, and Canada from 1997 to 2010. On average, $PM_{2.5}$ in Europe has been a little higher than in the US, while in Canada it has been one-third as much (Table 1 and Fig. 2). Generally, urban and suburban areas have similar $PM_{2.5}$ values, while values in rural areas are much less. Their variations (Fig. 6) are consistent with those of $PM_{10}$ (Fig. 4), but with amplified inter-annual variability. An increase in forest fires contributed to the peak values in 2003 in Europe (Trigo et al., 2006) and in 2005 in the US (Westerling et al., 2006).

The duration of the $PM_{2.5}$ data is shorter than that of $PM_{10}$ and there are no $PM_{2.5}$ data available for China. For the period when both $PM_{10}$ and $PM_{2.5}$ are available, linear trends of $PM_{2.5}$ are −9 % per decade (Europe), −18 % per decade (US), and −8 % per decade (Canada), while $PM_{10}$ has trends of −15 % per decade (Europe), −23 %





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per decade (US), and −7 % per decade (Canada). The decreasing trends of $PM_{2.5}$ in Europe and the US are substantially less than those of $PM_{10}$. As discussed below, finer particles, estimated from visibility measurements, have an opposite long-term trend to that of $PM_{10}$.

### 3.3 Contrasting trends of mass and optical properties of aerosols

The variations of $PM_{10}$ and optical extinction of aerosols in Europe and the US are compared using $PM_{10}$ sites where meteorological visibility has been measured less than 10 km away. For the cities in China, we paired the $PM_{10}$ and visibility using the city name because the Air Pollution Indices (APIs) are averaged from many sites in a city.

At monthly scales, $PM_{10}$ and optical extinction were closely correlated (Figs. 7 and 8). Both $PM_{10}$ and optical extinction have the largest seasonal variations in China. Therefore the correlation coefficients between $PM_{10}$ and optical extinction are the strongest in China. The concentration of $PM_{10}$ in the US and in Canada is very low as also optical extinction and both have low seasonal variations (Table 1).

The long-term trends of $PM_{10}$ and optical extinction are opposite in China, the US, and Canada (Fig. 9). From 1993 to 2010, optical extinction increased by 7 % over the US and by 10 % over Canada in spite of a substantial reduction of $PM_{10}$ in these two regions. Wild-fires in the US have increased during recent decades (Westerling et al., 2006). The smoke from the increased wild fires may explain the positive trend of optical extinction of aerosols in the US as the fine smoke particles produce large extinction but a very small contribution to the $PM_{10}$ mass concentrations.

The reduction of optical extinction over Europe of 5 % is also much less than the 44 % reduction in $PM_{10}$. Figure 9 shows that optical extinction of aerosols increased by 20 % in China from 2000 to 2005, and then decreased slightly, resulting in an overall increase of 18 % from 2000 to 2011. This increase in China is consistent with the increase of $SO_2$ (an important precursor of fine particles) estimated from satellite retrievals (Wang et al., 2010; Lyapustin et al., 2011; Itahashi et al., 2012), ground-based measurements (Lu et al., 2010), and model simulations (Lu et al., 2010). In particular,





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satellite-derived aeorosol optical depth (Fig. 4a of Itahashi et al., 2012) and modeled $SO_2$ emission (Fig. 2 of Lu et al., 2010) show nearly the same variability of optical extinction of aerosols derived from visibility. The cessation of the rapid increase since 2005 has been attributed to the application of flue-gas desulfurization devices in power plants in response to new Chinese policy (Lu et al., 2010). Several of China's mega-cites announced other measures (Wang and Chen, 2010) to control air pollution after 2005.

### 3.4 Precipitation impacts on mass and optical properties of aerosols

Small particles are primarily removed by wet deposition (precipitation), whereas large particles more by dry deposition (i.e., gravitational settling). Figure 9 shows that the variation in the number of precipitation days agrees more with the long-term variation of optical extinction than that of $PM_{10}$, as expected from these removal processes. Dry days are defined as the number of days without effective precipitation (rain gauge reading being larger than 0.1 mm) in a year. Both precipitation and visibility were observed at weather station. We paired them with $PM_{10}$ station using the same method discussed in Sect. 3.3. More numerous dry days imply less frequent removal of aerosols by rainfall as well as greater risk of wildfires, both of which result in higher aerosol concentrations in the atmosphere. The inter-annual variations of optical extinction over China and Europe agree well with the occurrence of dry days, with averaged correlation coefficients of 0.93 and 0.66, partly because optical extinction as well as $PM_{10}$ have large inter-annual variability (Fig. 10). Dry days vary less in the US and Canada (Fig. 10) and their contribution to the variation of aerosols is expected to be small. Inter-annual variation of optical extinction is consistent with the occurrence of dry days in the US however, the agreement of $PM_{10}$ and number of dry days is better in Canada (Fig. 10).





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## 4 Conclusions

This study compares the long-term variation of mass and optical properties of atmospheric aerosols over the Northern Hemisphere, including China, the US, Canada and Europe. Contrasting trends of mass and optical properties were found from 1992 to 2011. In particular, this synthesis of observational data on the mass and optical properties of aerosols from Europe, the US, Canada, and China over the past two decades shows that $PM_{10}$ has decreased substantially over Europe, the US, and Canada from 1992 to 2010 and over China's urban areas from 2000 to 2011. The duration of the $PM_{2.5}$ data is less than for $PM_{10}$, but when both $PM_{2.5}$ and $PM_{10}$ data were available, the decreasing trends of $PM_{2.5}$ were substantially less than those of $PM_{10}$ in Europe and the US. In addition, optical extinction increased in the US and Canada. It increased by 20 % over China's urban areas from 2000 to 2005 then slightly decreased.

There are numerous independent studies supporting the long-term trend of optical extinctions of atmospheric aerosols derived from meteorological visibility observations. In particular, satellite-derived AOD (Fig. 4a of Itahashi et al., 2012) and modeled $SO_2$ (Fig. 2 of Lu et al., 2010) show nearly the same variability of optical extinction of aerosols derived from visibility in China. The cessation of the rapid increase since 2005 has been attributed to the application of flue-gas desulfurization devices in power plants in response to new Chinese policy (Lu et al., 2010). The long-term variability of optical extinction derived from visibility has been confirmed by numerous independent estimates of atmospheric aerosols using direct measurement (Mitchell et al., 2010; Yoon et al., 2012), satellite retrievals (Lau and Kim, 2010; Dey and Di Girolamo, 2011), radiation measurements (den Outer et al., 2010; Ruckstuhl et al., 2010; Dwyer et al., 2010), and global chemical model simulations (Pozzoli et al., 2011; Chiacchio et al., 2011).

These results indicate that although mass concentration of atmospheric aerosols has substantially decreased, optical extinction of aerosols has either decreased less or has increased, i.e., fine particles increased or decreased less as optical extinction





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is sensitive to particles of sizes less than 1 µm. The number of these particles can be large, and they are especially important for their impact on human health (Fenger, 2009).

The reason for the increase of optical extinction (or atmospheric aerosols) has been proposed as more anthropogenic emission of atmospheric aerosols (Myhre, 2009), increased forest fires (Yoon et al., 2012), and more fossil fuel emission from developing countries, such as China and Indian (Wang et al., 2009). These fine particles account for a small part of total mass of aerosols. However, they are very efficient in light extinction. Evidently, the ratio of efficiently-scattering atmospheric particles (Hand and Malm, 2007; Hoff and Christopher, 2009) to total particles has increased in all of the regions studied. A recent study consistent with our results shows that the relative increase in anthropogenic black carbon is much larger than the overall increase in the anthropogenic abundance of aerosols (Myhre, 2009). This increase is important for climate, affecting the global circulation and hydrological cycle (Menon et al., 2002; Ramanathan et al., 2007).

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**Table 1.** Multi-year regional averages of $PM_{10}$ and $PM_{2.5}$ (unit: $\mu g\,m^{-3}$) with a standard deviation in the study regions.

| | Region (time period) | Total | Urban | Suburban | Rural |
|---|---|---|---|---|---|
| **$PM_{10}$** | China (2000–2011) | | 96.5 ± 26.4 | | |
| | Europe (1992–2009) | 28.9 ± 8.6 | 30.6 ± 8.4 | 28.3 ± 7.5 | 24.0 ± 9.2 |
| | USA (1993–2010) | 27.0 ± 7.9 | 27.6 ± 8.1 | 26.2 ± 7.2 | 28.4 ± 9.8 |
| | Canada (1993–2009) | | 15.5 ± 5.2 | | |
| **$PM_{2.5}$** | Europe (1998–2009) | 15.8 ± 6.5 | 17.5 ± 5.9 | 17.8 ± 4.7 | 11.3 ± 6.6 |
| | USA (1998–2010) | 12.2 ± 3.5 | 12.5 ± 3.5 | 12.7 ± 3.2 | 10.0 ± 3.4 |
| | Canada (1995–2009) | 5.6 ± 2.3 | 5.6 ± 1.7 | | 3.8 ± 1.5 |





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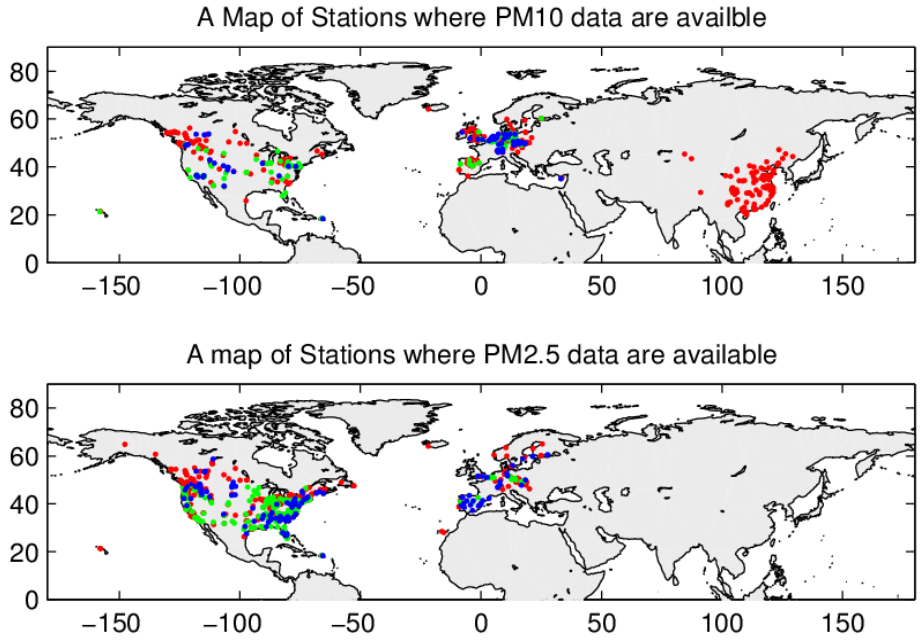


**Fig. 1.** (Top panel) A map of stations over China, Europe, US, and Canada (red points: urban sites, green points: suburban sites, and blue points: rural sites). Europe and US stations have more than 120 months of data of $PM_{10}$ during the study period, while China and Canada stations have more than 60 months of data of $PM_{10}$ available during the study period. (Bottom panel) A map of stations over Europe, the US, and Canada (red points: urban sites, green points: suburban sites, and blue points: rural sites) where $PM_{2.5}$ data are available for more than 60 months from 1998 to 2010.





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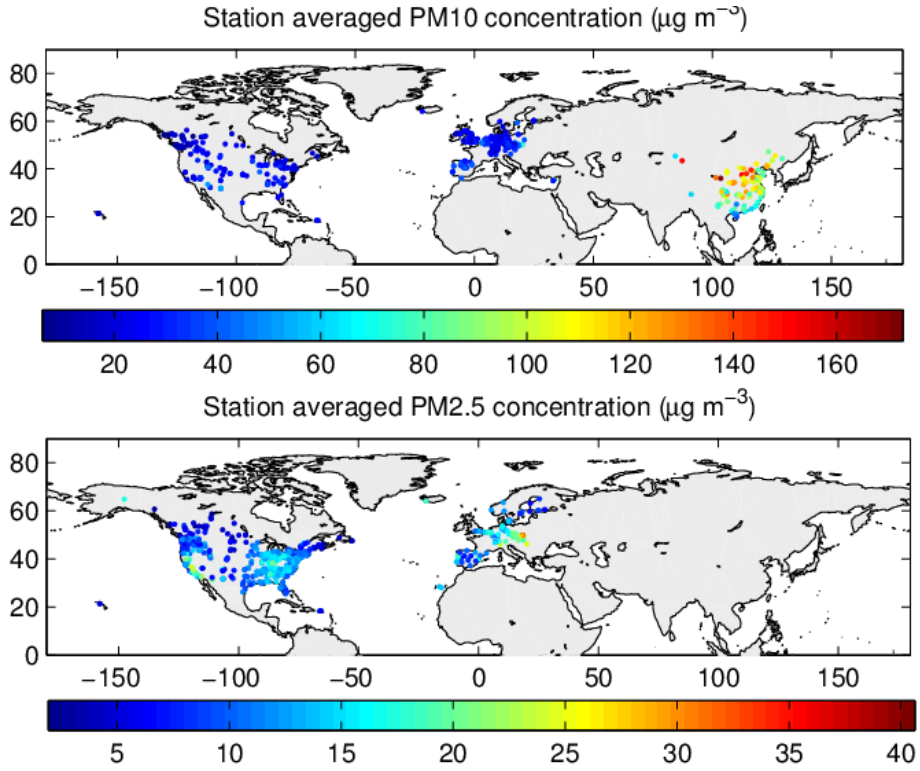


**Fig. 2.** A map of station multi-year averaged $PM_{10}$ and $PM_{2.5}$ mass concentration (unit: $\mu g\,m^{-3}$).





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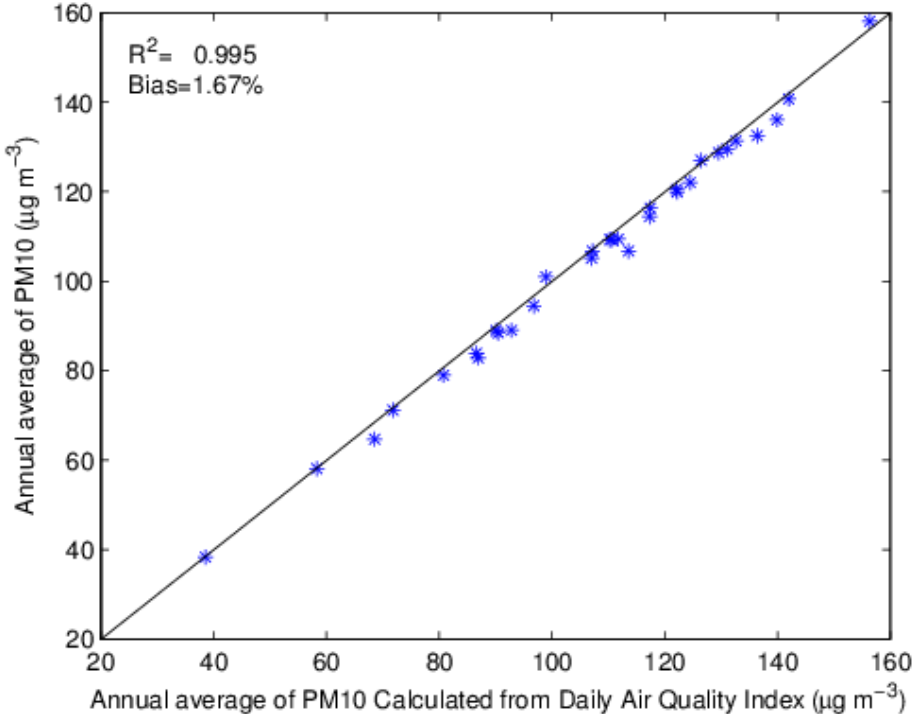


**Fig. 3.** The comparison of site-average $PM_{10}$ calculated from Air Quality Index and those averaged from $PM_{10}$ measurements from 2003 to 2009 in 31 major cities in China.





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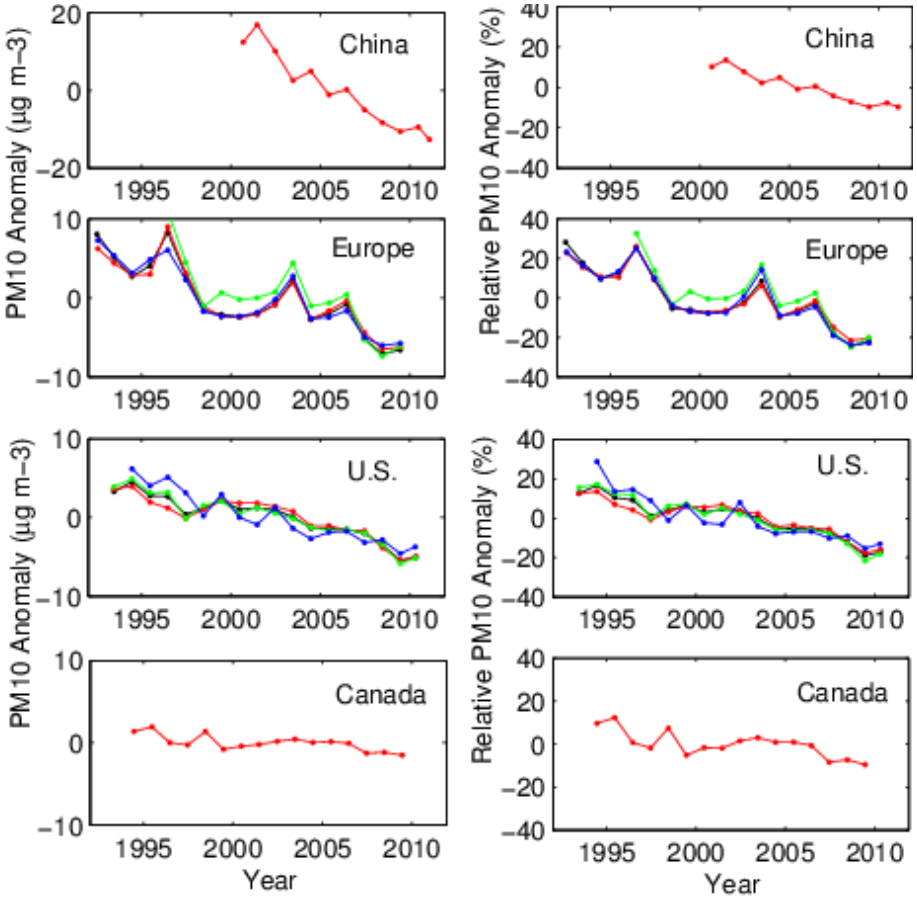


**Fig. 4.** Annual anomalies (left) of $PM_{10}$ and their relative anomalies (normalized by multi-year averaged $PM_{10}$ value from each site) (right). Black lines represent averages over total sites, red lines represent urban averages, green lines represent suburban averages, and blue lines represent rural averages. Multi-year regional averages of $PM_{10}$ can be found in Table 1.





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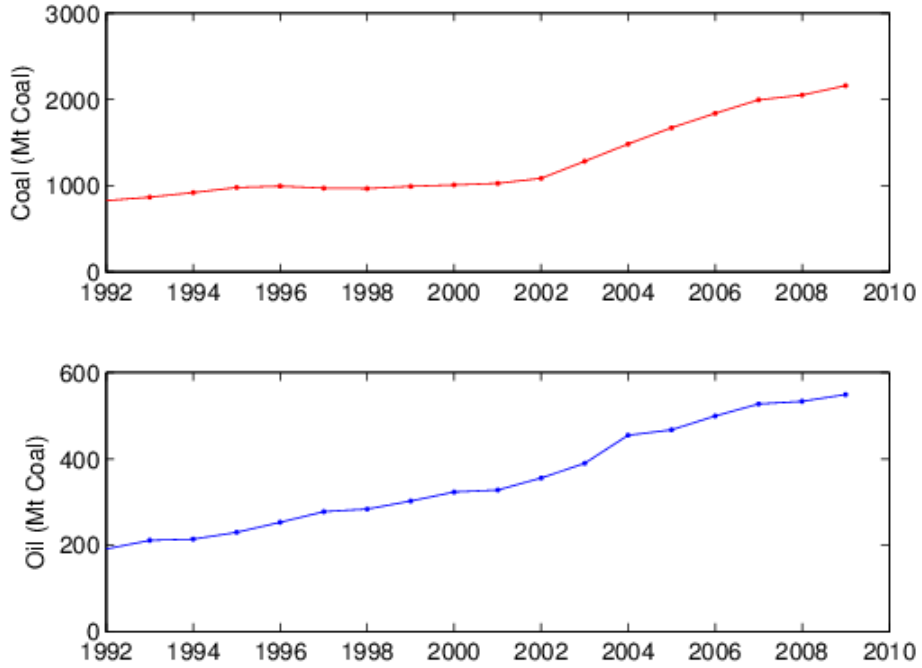


**Fig. 5.** Total coal and oil consumption in China from 1992 to 2009 (unit: million ton coal, data source: National Bureau of Statistics of China, available at http://www.stats.gov.cn/tjsj/ndsj/, in Chinese).





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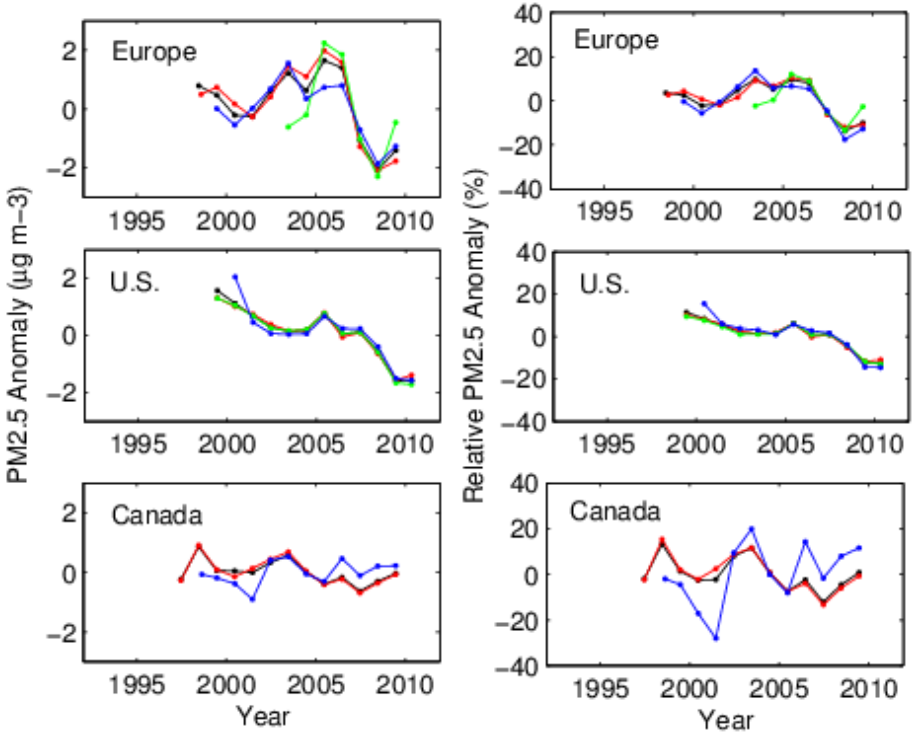


**Fig. 6.** Annual anomalies (left) of $PM_{2.5}$ and their relative anomalies (normalized by multi-year-averaged $PM_{2.5}$ value from each site). Black lines represent averages over total sites, red lines represent urban averages, green lines represent suburban averages, and blue lines represent rural averages. Multi-year regional averages of $PM_{2.5}$ can be found in Table 1.





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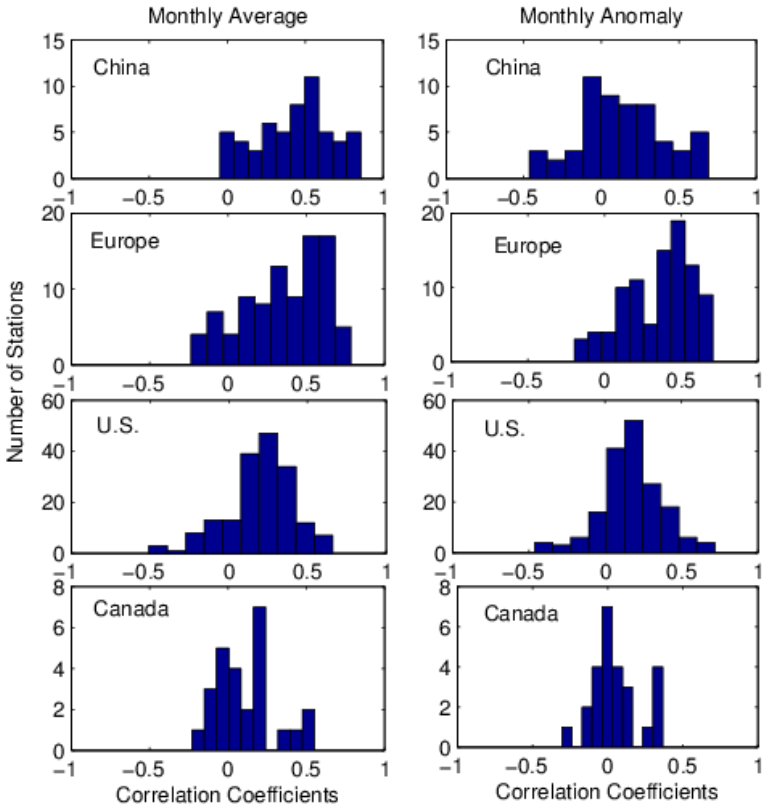


**Fig. 7.** Left column: the histograms of correlation coefficients, computed at each station, between monthly averages of $PM_{10}$ and optical extinction coefficients (inverse of visibility); right column: same to left column except for the correlation coefficients between monthly anomalies (seasonal cycle removed). Regions are as indicated in each panel. Data used here are the same as those used in Figs. 9 and 10.





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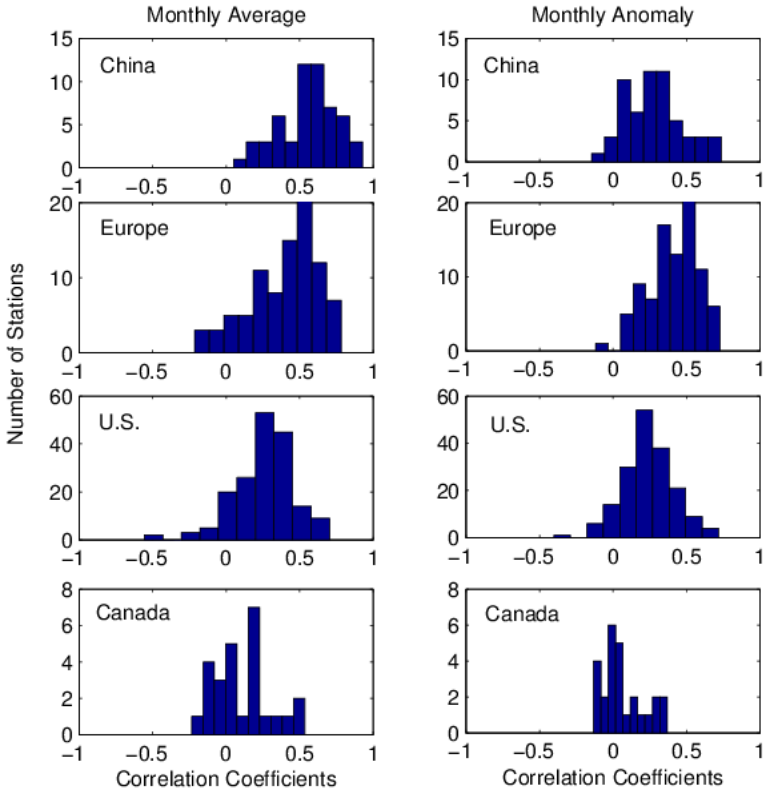


**Fig. 8.** Same as Fig. 7 except that the long-term trends were removed from the monthly averages and monthly anomalies (seasonal cycle removed) of $PM_{10}$ and optical extinction coefficients (inverse of visibility).





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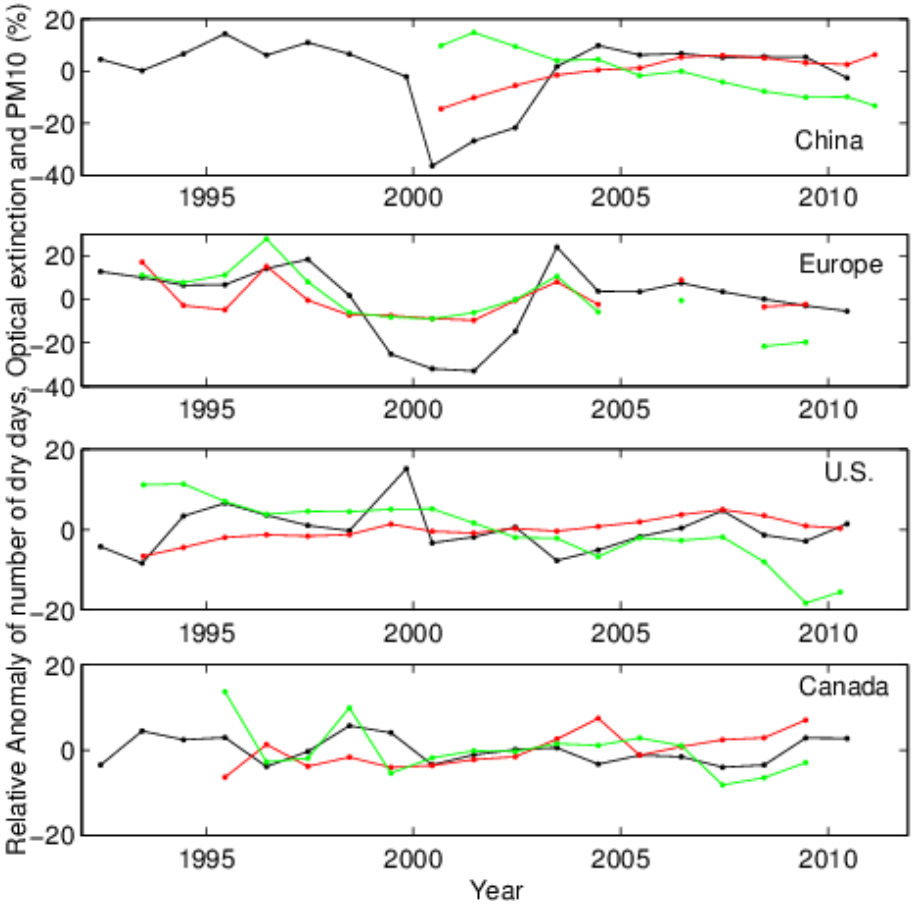


**Fig. 9.** Annual anomaly of normalized number of dry days (black), optical extinction coefficients (inverse of visibility, red), and $PM_{10}$ (green) averaged from sites where $PM_{10}$ data are available for more than 120 months for Europe and the US and for more than 60 months for China and Canada.





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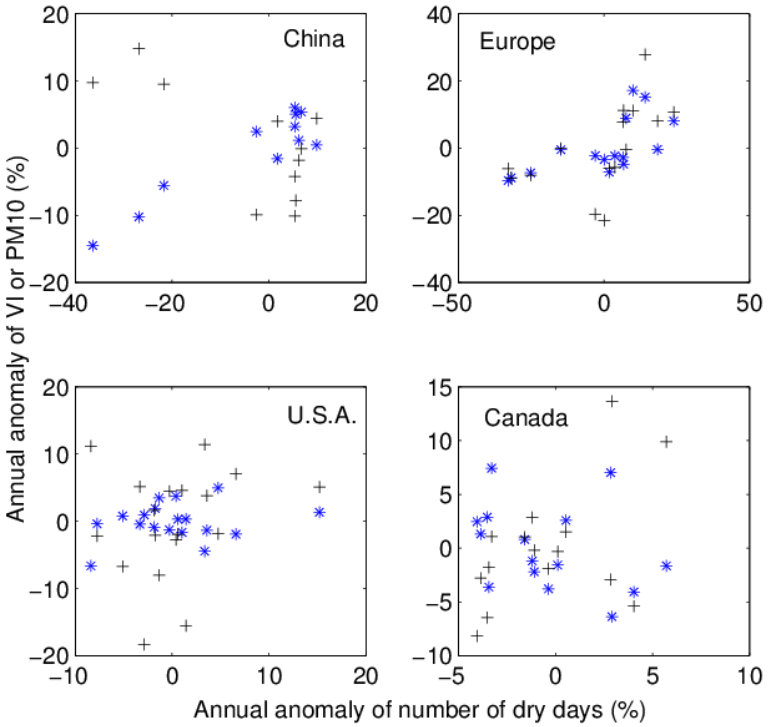


**Fig. 10.** Scatterplots of regional averaged annual anomaly of aerosol optical extinction (inverse of visibility, blue star) and $PM_{10}$ (Black plus) as a function of annual anomaly of number of dry days in China, Europe, the US, and Canada. Data used are same to Fig. 3. The correlation coefficients between optical extinction and dry days are 0.93 (China), 0.66 (Europe), 0.23 (the US) and −0.32 (Canada). The correlation coefficients between $PM_{10}$ and dry days are −0.72 (China), 0.55 (Europe), 0.20 (the US) and 0.53 (Canada).





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