A point-by-point response to the reviews

We thank the reviewers for the constructive comments. Below we present the comments of the reviewers (bold) and our replies with the proposed modification to the manuscript. In addition to the changes, we discovered a minor error in the calculations during the revision of the manuscript. While the scientific contents and our conclusions do not change, we now estimate a slightly lower number of mortality attributable to air pollution in Beijing (around 10%).

Reply to the comments made by Reviewer #3

1. As described in the paper, the yearly premature morality (Fig. 6) is calculated from the yearly average of $PM_{2.5}$ and yearly registered population (the latter is presented in Fig.5). Although day-on-day $PM_{2.5}$ from 2001 to 2012 are shown in Fig. 2, the trend of $PM_{2.5}$ cannot be seen clearly from the figure. I would suggest that the yearly averaged $PM_{2.5}$ from 2001 to 2012 be presented and its variation on the estimated trend of premature morality discussed.

We have followed the suggestions, and show the yearly averaged $PM_{2.5}$ from 2001 to 2012 in a new figure. From the figure we can infer that there is a positive trend in the estimated AOD trend in Beijing between 2001-2012, although no clear increase is present after the year 2004.

2. As shown in Fig. 1, there could be large biases in daily $PM_{2.5}$ estimated from AOD. How about the uncertainties in estimated yearly averaged $PM_{2.5}$? It might not be so large as for the daily averaged values, but a quantitative estimate is needed. For example, the yearly averaged $PM_{2.5}$ estimated from AOD for the year 2012 can be compared with that calculated from the original $PM_{2.5}$ measured at the embassy site.

We have compared the estimated $PM_{2.5}$ to the ground-based $PM_{2.5}$, also we have compared the estimated $PM_{2.5}$ to measured $PM_{2.5}$ in Beijing from previous studies. In our study, the average estimated $PM_{2.5}$ from May 10, 2010 to December 6, 2011 is 104 $\mu g/m^3$. The average ground-based observed $PM_{2.5}$ is 93 $\mu g/m^3$, being rather close. Han et al. (2007) investigated $PM_{2.5}$ concentrations in Beijing from 2001 to 2004 and found during summer of 2002, spring and autumn of 2003 that it was 79.6 $\mu g/m^3$, 111.6 $\mu g/m^3$ and 107.3 $\mu g/m^3$, respectively. In this study, the estimated $PM_{2.5}$ during summer of 2002, spring and autumn of 2003 was 73.7 $\mu g/m^3$, 99.9 $\mu g/m^3$, and 78 $\mu g/m^3$. Wang et al. (2009) found in summer and winter during 2005-2007 in Beijing that the average $PM_{2.5}$ was 102 $\mu g/m^3$. During the same period, our estimated $PM_{2.5}$ was 99 $\mu g/m^3$. We can summarize that our calculations are well within 10% of the observed values present in the literature.

3. The authors state that ground-based $PM_{2.5}$ is not available for the period 2001-2012 in Beijing (P28660, Line 24-25). This might be true for long-term CONTINUOUS measurements of $PM_{2.5}$ over this period. In fact, there have been numerous measurements of $PM_{2.5}$ as well as its chemical components in Beijing since the

early 2000s, as reported in both domestic and international publications including some review pa-pers. The authors may consider the possibility of using these observational data to validate their estimated $PM_{2.5}$ for specific years or seasons.

We have revised that sentence. Also, we have compared the estimated $PM_{2.5}$ to measured $PM_{2.5}$ from previous studies in Beijing for specific seasons (see previous reply).

Reply to the comments made by R.P. Singh

1. **The approach of Zheng et al. is interesting but it has uncertainties in estimating $PM_{2.5}$ and yearly premature mortality. The sources of uncertainties must be discussed by the authors.**

Uncertainties in the method applied here have been already partially discussed in Lelieveld et al. (2013). However, we added in the manuscript a discussion on uncertainties. It must however be stressed that we included only uncertainties from the Concentration-Response function. This, in fact, is associated with the highest uncertainties, typically much larger than from $PM_{2.5}$ and AOD measurements. Finally, it is difficult (if not impossible) to quantify the uncertainties derived from the population database used, which is anyhow discussed in the manuscript.

2. The paper is interesting and important to bring out the attention of people about the poor air quality and its direct impact on human health and increasing mortality rate so that the sources of air pollution is reduced. Various sources of pollution in Beijing city may be added in the paper.

Sun et al. (2004), based on aerosol samples from 2002 to 2003 in Beijing, showed that coal burning and traffic exhausts, plus the dust through long-range transport, could be the major sources of the aerosol pollution in Beijing. The winter heavy fog in Beijing is not only correlated with local pollution emission, but also with long distance pollution transport from the surrounding areas of Beijing, such as Tianjin city, Hebei and Shandong provinces, etc. (Ma et al., 2010; Shi and Xu, 2012). Zhang et al. (2013), based on 121 daily $PM_{2.5}$ samples collected in Beijing, showed that soil dust, coal combustion, biomass burning, traffic and waste incineration emission, industrial pollution, and secondary inorganic aerosol are the six main sources of $PM_{2.5}$ aerosol speciation, and demonstrated that regional sources could be crucial contributors to PM pollution in Beijing.

3. The authors may consider to show a high resolution map of Beijing with Aeronet and US Embassy locations where air quality data was monitored. Air quality data ($PM_{2.5}$) considered in the recent study may be shown and its daily variations may also be discussed.

Following suggestions of the Referee, we show a map of Beijing with AERONET and U.S. Embassy locations, as well as administrative divisions including Beijing central area defined in the study in Fig. 1. We have discussed the seasonal variability in $PM_{2.5}$ concentration, and compared it to previous studies in the part “4.2 Correlation analysis”.

4. In Figure 2, authors may consider to show AOD variations.

Following suggestions of the Referee, we show AOD variations in Fig. 2.

5. The authors may discuss the importance of BLH and RH which are important parameters in the dynamics of atmospheric pollutants and also in weather conditions.

The correlation between AOD and $PM_{2.5}$ is strongly influenced by the vertical distribution of aerosols and the RH that impacts aerosol extinction coefficient. These two factors are related to atmospheric profiles, ambient conditions, as well as the size distributions and chemical compositions of aerosols, and they may have large spatial and temporal variations (Wang et al., 2010).

6. It will be interesting if the authors can show different age group of people who suffer with the increasing $PM_{2.5}$.

We have revised the figures, showing the yearly premature mortality by IHD, CEV, COPD and LC for people >30 yrs, and ALRI for infants <5 yrs from 2001 to 2012

Reply to the comments made by Reviewer #5

One major concern of this paper is the uncertainty. Instead of point estimate, it is more encouraged that the authors present the range the uncertainty (or confidential interval).

We understand the comments of the referee. To have a sound discussion of the uncertainties, we have compared the estimated $PM_{2.5}$ to the ground-based $PM_{2.5}$, also from previous studies for specific seasons. The mortality estimations have been estimated also for the upper-lower limit based on the Concentration-Response function confidence interval (95% confidence level) as these uncertainties are by far the largest in the calculations.

Specific comments:

1. Page 28660, Lines 12-13, authors need to add citation support to the statement "AOD and PM are related to atmospheric profiles, ambient conditions, as well as the chemical composition of aerosols". Besides, there is no incorporation of these parameters in the development of AOD-$PM_{2.5}$ equation except BLH and RH. Are these parameters represented by BLH and RH? If then, the authors may need to reorganize the context to make this clear. The authors mentioned twice that the relationship between AOD and $PM_{2.5}$ is influenced by the chemical components of particles, both on Page 28660 Line 13 and on page 28664 line21. However, this parameter was not incorporated into the AOD- $PM_{2.5}$ equation; neither is discussed regarding its impact on the uncertainty of the results. Further elaboration on this statement is needed.

We have deleted this sentence, and correct the statement and cite one reference in the part "4.1 Influence of the BLH and ambient RH". The correlation between AOD and $PM_{2.5}$ is

strongly influenced by the vertical distribution of aerosols and the RH that impacts aerosol extinction coefficient. These two factors are related to atmospheric profiles, ambient conditions, as well as the size distributions and chemical compositions of aerosols, and they may have large spatial and temporal variations (Wang et al., 2010). In order to reduce the uncertainties, the atmospheric BLH and ambient RH have been introduced into the correlation analysis.

2. Page 28660, lines 13-25, the time series method in the epidemiological studies and the study conducted in this paper are two different kinds of research. The former one is to develop the C-R function from known $PM_{2.5}$ concentration and mortality data, while the latter uses the developed C-R function and PM concentration to estimate the mortality. It is confusing to use the comparison these two research as a motivation of this paper.

The time series method and the study conducted in this paper are two different kinds of research. The C-R function is based on epidemiological cohort studies. There is no relationship between the time series method and the method in this study, and we do not intend to compare these two researches. The sentence about the time series method is used to conclude previous studies regarding the relationship between PM concentrations and mortality in Beijing. Then we find the limitation of these studies, and introduce our method.

3. Page 28662, Line 9, why AOD at wavelength 550nm is selected to derive the ground-level $PM_{2.5}$?

Since most satellite AOD is derived at wavelength 550nm, and most atmospheric models also adopt this band, we select AOD at wavelength 550nm for the better comparison with the correlation coefficient between AOD (550nm) and $PM_{2.5}$ from previous studies.

4. Page 28665, Line 3, it says "g is an empirical fit coefficient, and it equals 1 in this study." What is the evidence for this value to be set to 1?

The coefficient g is an empirical fit coefficient. This value is set to 1 following previous studies in Beijing. Generally, RH influencing factor could be expressed as follows: $f(RH)=1/(1.0-RH/100)$. A more accurate correction could be obtained from experiments (Li et al., 2005).

5. Page 28662, the seasonal distribution and characteristics of AOD was presented. How does it affect the estimation of the ground-level $PM_{2.5}$ concentration and mortality caused? The seasonal variability in PM concentration and mortality should also be discussed.

We have discussed the seasonal variability in $PM_{2.5}$ concentration in the part "4.2 Correlation analysis". For the monthly data, the highest mean of AOD and $PM_{2.5}$ does not occur in the same month, since the relationship between AOD and ground-level $PM_{2.5}$ is affected by Boundary Layer Height (BLH) and Relative Humidity (RH) in our model. Because long-term exposure to $PM_{2.5}$ is associated with increased mortality, we discussed the yearly mortality due to $PM_{2.5}$, and no seasonal variability of mortality is discussed. Also the

yearly mortality is affected by yearly $PM_{2.5}$ concentration, population, and baseline mortality rate.

References

Han, L.H., Zhuang, G.S., Cheng, S.Y., and Li, J.: The mineral aerosol and its impact on urban pollution aerosols over Beijing, China, Atmos. Environ., 41, 7533-7546, 2007.

Lelieveld, J., Barlas, C., Giannadaki, D., and Pozzer, A.: Model calculated global, regional and megacity premature mortality due to air pollution, Atmos. Chem. Phys., 13, 7023-7037, doi:10.5194/acp-13-7023-2013, 2013.

Li, C.C., Mao, J.T., Lau, A.K.H., Yuan, Z.B., Wang, M.H., and Liu, X.Y.: Application of MODIS satellite products to the air pollution research in Beijing, Sci. China Ser. D, 48, Supp. II, 209-219, 2005

Ma, J.Z., Chen, Y., Wang, W., Yan, P., Liu, H.J., Yang, S.Y., Hu, Z.J., and Lelieveld, J.: Strong air pollution causes widespread haze-clouds over China, J. Geophys. Res., 115, D18204, 2010.

Shi, X.H., and Xu, X.D.: Progress in the study of regional impact of aerosol and related features of heavy fog in Beijing City (In Chinese), Chinese Journal of Geophysics, 55, 3230-3239, 2012.

Sun, Y.L., Zhuang, G.S., Wang, Y., Han, L.H., Guo, J.H., Dan, M., Zhang, W.J., Wang, Z.F., and Hao, Z.P.: The air-borne particulate pollution in Beijing-concentration, composition, distribution and sources, Atmos. Environ., 38, 5991-6004, 2004.

Wang, H.L., Zhou, Y.M., Zhuang, Y.H., Wang, X.K., and Hao, Z.P.: Characterization of $PM_{2.5}$/$PM_{2.5\text{-}10}$ and source tracking in the juncture belt between urban and rural areas of Beijing, Chin. Sci. Bull., 54, 2506-2515. 2009.

Wang, Z.F., Chen, L.F., Tao, J.H., Zhang, Y., and Su, L.: Satellite-based estimation of regional particulate matter (PM) in Beijing using vertical-and-RH correcting method, Remote Sens. Environ., 114, 50-63, 2010.

Zhang, R., Jing, J., Tao, J., Hsu, S.-C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, doi:10.5194/acp-13-7053-2013, 2013.

A list of all relevant changes made in the manuscript

We appreciated the editor's work and the three reviewers' efforts. We have read the reviews carefully and revised the manuscript following their comments/suggestions. The specific changes we have done to the original manuscript are listed as followings.

In the title part, we have changed it to "Long-term (2001-2012) concentrations of fine particulate matter ($PM_{2.5}$) and the impact on human health in Beijing, China".

In the abstract part, we have revised the average total mortality and the per capita mortality, since we have corrected a minor error in the calculations.

In the part "1 Introduction", following the reviewers' comments, we have added various sources of pollution in Beijing, and added the corresponding references. We have also revised some sentence to make it concise. For example, the sentence "AOD and PM are related to atmospheric profiles, ambient conditions, as well as the chemical composition of aerosols" has been deleted, and the sentence "ground-based $PM_{2.5}$ is not available for the period 2001-2012 in Beijing" has been changed to "long-term continuous measurements of $PM_{2.5}$ for the period 2001-2012 in Beijing are not available".

In the part "2 Data", following the reviewers' comments, we have added the sentence about the map of Beijing, and also show the map in Fig. 1.

In the part "3 Analyzing AERONET AOD", following the reviewers' comments, we have added the sentence about the AOD at 550 nm wavelength and show its variation in Fig. 2. In addition, we have changed the sentence "Winter (December-February) is dominated by cold, dry, and windy weather due to cold air from the west Siberian anticyclone" to "Winter (December-February) is dominated by cold, dry, and windy weather due to cold air advected by the west Siberian anticyclone".

In the part "4.1 Influence of the BLH and ambient RH", following the reviewers' comments, we have added a sentence to describe the importance of BLH and RH, and cited one reference. Also, in order make the content concise, the sentence "The correlation between aerosol extinction coefficient and $PM_{2.5}$ concentration is influenced by the chemical components of particles and RH of the ambient air" has been deleted.

In the part "4.2 Correlation analysis", we have revised the figure number, since we have added some figures. We have followed the reviewers' suggestions, and added the sentence about the yearly averaged $PM_{2.5}$ from 2001 to 2012. We have compared the estimated $PM_{2.5}$ to the ground-based $PM_{2.5}$, also we have compared the estimated $PM_{2.5}$ to measured $PM_{2.5}$ in Beijing from previous studies.

In the part "5.1 Beijing central area", we have revised the figure number.

In the part "5.2 Concentration-response functions", we have deleted the description of the Integrated Exposure Response (IER) model, since we have revised the calculation.

In the part "5.3 Mortality estimation and discussion", we have revised the calculation method, i.e., "we used the relationship between $PM_{2.5}$ and RR which are organized in bins from global burden of disease study 2010 (GBD 2010)". We have deleted the description related to the IER model, and revised the equation number. Also, we have added some discussion about the per capita mortality for the period 2010-2012. Following the reviewers'

comments, we have added the descriptions of the uncertainties of the mortality estimation. Since we have revised the calculation method, we have revised the premature mortality and the per capita mortality, and added the corresponding uncertainties. Additionally, we have compared the estimation of the mortality due to $PM_{2.5}$ with past studies, and added the corresponding references.

In the part "6 Conclusion", we have revised the average total mortality and the per capita mortality, and added the uncertainties. In addition, we have added the assumption "If we assume the range of our and previous studies to be representative of the city of Beijing, this implies a mortality attributable to $PM_{2.5}$ of about 22,000 - 30,000 persons per year", and revised some sentence to make it clear.

In the part "Acknowledgements", we have added one word "the".

In the part "References", we have added and deleted some references. For example, the reference "Burnett et al., 2014" has been deleted, and the references "Ma et al., 2010; Wang et al., 2009; Zhang et al., 2013" have been added.

In Table 1, we have revised the calculation results, since we have revised the calculation method. Also, we have added the uncertainty ranges of the total mortality estimation

In the figures part, following the reviewers' comments, we have added Fig. 1, Fig. 2, and Fig 4(b), and revised the figure of the annual premature mortality. We have also revised the descriptions of the figures.

A marked-up manuscript

Long-term (2001-2012) concentrations of fine particulate matter ($PM_{2.5}$) and the impact on human health in Beijing, China

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Abstract

Beijing, the capital of China, is a densely populated city with poor air quality. The impact of high pollutant concentrations, in particular of aerosol particles, on human health is of major concern. The present study uses Aerosol Optical Depth (AOD) as proxy to estimate long-term $PM_{2.5}$, and subsequently estimates the premature mortality due to $PM_{2.5}$. We use the AOD from 2001 to 2012 from the Aerosol Robotic Network (AERONET) site in Beijing and the ground-based $PM_{2.5}$ observations from the U.S. embassy in Beijing from 2010 to 2011, to establish a relationship between $PM_{2.5}$ and AOD. By including the atmospheric boundary layer height and relative humidity in the comparative analysis, the correlation (R^2) increases from 0.28 to 0.62. We evaluate 12 years of $PM_{2.5}$ data for the Beijing central area using an estimated linear relationship with AOD, and calculate the yearly premature mortality by different diseases attributable to $PM_{2.5}$. The estimated average total mortality due to $PM_{2.5}$ is about ~~6,100~~ 5,100 individuals/yr for the period 2001-2012 in the Beijing central area, and for the period 2010-2012 the per capita mortality for all ages due to $PM_{2.5}$ is around 15~~17.9~~ per 10,000 person-year, which underscores the urgent need for air pollution abatement.

1 Introduction

Air pollution has intensified strongly since the industrial revolution, i.e., during the epoch known as the Anthropocene (Crutzen, 2002). Ground-level fine particulate matter with a diameter <2.5 μm ($PM_{2.5}$) has increased substantially, not only in most urbanized and industrialized areas but also in rural and even remote regions (Akimoto, 2003; Anenberg et al., 2010; Schulz et al., 2006). Aerosols have extensive impacts on our climate and environment (Kaufman et al., 2002). $PM_{2.5}$ can have serious health impacts by cardiovascular and respiratory disease and lung cancer, and especially chronic exposure is associated with morbidity and premature mortality (Dockery et al., 1993; McDonnell et al., 2000; Pope III et al., 2009). Concentration-response functions have been used to estimate mortality due to $PM_{2.5}$ from anthropogenic sources. Globally, air pollution has been estimated to represent a significant fraction~~1.4%~~ of the total mortality attributable to 26 risk factors assessed by the World Health Organization (WHO) global burden of disease project (GBD) (Ezzati et al., 2002). Cohen et al. (2004) estimated that urban $PM_{2.5}$ exposure is responsible for approximately 712,000 cardiopulmonary disease (CPD) and 62,000 lung cancer deaths in 2000. Anenberg et al. (2010) found that anthropogenic $PM_{2.5}$ is associated with 3.5 million CPD and 220,000 lung cancer mortalities annually. Evans et al. (2012) undertook a global assessment of mortality associated with long-term exposure to fine particulate air pollution using remote sensing data and found that the global fraction of adult mortality attributable to the anthropogenic component of $PM_{2.5}$ is 8.0% for CPD and 12.8% for lung cancer. The GBD for 2010 indicates that outdoor air pollution in the form of fine particles is a much more significant public health risk than previously assumed (Lim et al., 2012). In China, the GBD estimates 1.2 million premature deaths. Outdoor air pollution ranks number 4 among leading risk factors contributing to deaths in China in 2010.

China has undergone very rapid economic growth since the economic reform beginning in 1978. This has resulted in an increase in energy consumption, air pollution and associated health problems (HEI International Oversight Committee, 2004). Beijing, as a megacity and the capital of China, is one of the most populous cities in the world with 20 million inhabitants (in 2011) over an area of 16,800 km^2. It faces serious air pollution and associated human health problems. Several studies on the characteristics of aerosols in Beijing have been carried out (Cao et al., 2002; Han et al., 2013; Sun et al., 2012; Winchester and Mu-Tian, 1984; Yang et al., 2000), showing that industrial emissions, vehicle exhausts, dust and coal burning are major causes of particulate pollution in Beijing. Sun et al. (2004), based on aerosol samples from 2002 to 2003 in Beijing, showed that coal burning and traffic exhausts, plus the dust through long-range transport, could be the major sources of the aerosol pollution in

Beijing. The winter heavy fog in Beijing is not only correlated with local pollution emission, but also with long distance pollution transport from the surrounding areas of Beijing, such as Tianjin city, Hebei and Shandong provinces, etc. (Ma et al., 2010; Shi and Xu, 2012). Zhang et al. (2013), based on 121 daily $PM_{2.5}$ samples collected in Beijing, showed that soil dust, coal combustion, biomass burning, traffic and waste incineration emission, industrial pollution, and secondary inorganic aerosol are the six main sources of $PM_{2.5}$ aerosol speciation, and demonstrated that regional sources could be crucial contributors to particulate matter (PM) pollution in Beijing. Regarding PM~~particulate matter (PM)~~ in Beijing, both PM_{10} and $PM_{2.5}$ have been extensively studied (Hu et al., 2013; Li et al., 2013b; Sun et al., 2006; Zhang et al., 2013; Zhao et al., 2013; Zhao et al., 2009; Zhu et al., 2011). The highest PM_{10} concentrations in Beijing typically occur in April and October according to the records from 2003 to 2009 (Zhu et al., 2011). From the daily PM_{10} concentration measurements collected at 27 stations in Beijing over a 5-year period, it is found that the overall trend of PM_{10} is generally negative, which applies in particular to summer and winter, while in spring the concentration has increased in recent years (Hu et al., 2013). Pronounced seasonal variation of $PM_{2.5}$, measured from 2005 to 2007 at 5-min time resolution, occurs in the urban area in Beijing, with the highest concentrations typically in the winter and the lowest in the summer (Zhao et al., 2009).

Satellite derived aerosol optical depth (AOD) and aerosol concentrations at the surface ($PM_{2.5}$, PM_{10}) have been analyzed, and high correlations have been found (Li et al., 2005; Gupta et al., 2006; Wang et al., 2010b; Zheng et al., 2014). These correlations are partly based on models to infer the surface data from column satellite data, and are strongly influenced by the assumed vertical distribution of aerosols and the relative humidity. ~~AOD and PM are related to atmospheric profiles, ambient conditions, as well as the chemical composition of aerosols.~~

Epidemiological research using time series methods has shown the relationship between PM concentrations and human health in Beijing associated with mortality and morbidity (Li et al., 2013a; Zhang et al., 2012a; Zhang et al., 2012b). Nevertheless, these studies have focused on particular periods of a few years or less. In addition, most of these epidemiological studies are based on limited ground-based $PM_{2.5}$ and PM_{10} measurements, which may not represent the city. Since 2013 the Beijing Municipal Environmental Monitoring Centre has started to publish $PM_{2.5}$ data and has included it in the calculation of air quality index (AQI) (Zheng et al., 2014). In addition to air pollution, the population in Beijing has steadily increased over the past decades, being 13 million in 2000 and growing to 21 million in 2013. The long term $PM_{2.5}$ and premature mortality estimation will support ~~help with~~ policy decisions aimed at reducing health impacts of $PM_{2.5}$. However, long-term continuous measurements of ~~ground-based~~ $PM_{2.5}$ for the period 2001-2012 in Beijing ~~is~~ are not available ~~for the period 2001-2012 in Beijing~~, let alone the premature mortality due to $PM_{2.5}$.

In the present study, we use AOD as proxy to estimate long term $PM_{2.5}$, and then estimate the premature mortality due to $PM_{2.5}$ to assess to what degree $PM_{2.5}$ affects human health in Beijing. We collect the long term AERONET AOD, and analyze its seasonal variability. A linear regression model for $PM_{2.5}$ has been established based on AOD, considering boundary layer height (BLH) and relative humidity (RH) corrections, allowing the reconstruction of $PM_{2.5}$ concentrations for the last decades. Furthermore, the annual premature mortality attributable to different diseases caused by $PM_{2.5}$ has been estimated by employing concentration-response functions based on epidemiological cohort studies.

2 Data

In this work we use ground-based $PM_{2.5}$, AOD and relative humidity (RH) observations from the U.S. embassy in Beijing, AERONET and the China Meteorological Data Sharing Service System, respectively. The map of Beijing with AERONET and $PM_{2.5}$ stations is shown in Fig. 1. We have adopted daily $PM_{2.5}$ data from the U.S. embassy monitoring station as published by Wang et al. (2013).

The U.S. embassy is located in the Chaoyang district. Hourly $PM_{2.5}$ concentrations are reported by the U.S. embassy and made available via the Internet. The U.S. embassy monitors the energy decay of beta rays to assess the concentration of particles in the atmosphere. The results obtained from beta ray measurements are usually at least 15 percent higher than those collected by oscillating microbalance, according to data on the website of the China National Environmental Monitoring Center (http://usa.chinadaily.com.cn/epaper/2012-10/30/content_15856991.htm). Wang et al. (2013) gathered $PM_{2.5}$ at the U.S. embassy station in Beijing from May 10, 2010, to December 6, 2011. Days with extended periods of missing $PM_{2.5}$ (hourly) data were discarded based on the following criterion: during a day there are consecutive data gaps of more than 3 hours or the cumulative amount of missing data exceeds 12 hours. The final dataset covers a 423-day period.

The AOD observations are obtained from the Aerosol Robotic Network (AERONET) program, which is a federation of ground-based remote sensing aerosol networks to measure aerosol optical properties (Holben et al., 1998). We use the AERONET level 2.0 data, which are cloud screened and quality assured. The AERONET data for the Beijing site starts on March 7, 2001, and ends on September 19, 2012, and encompasses the AOD at the four wavelengths 1020, 870, 675 and 500 nm. The AERONET data provides AOD in the form of all points, daily averages, and monthly averages. The daily average AOD is used in this study. The daily RH at the Beijing national meteorological station has been taken from the China Meteorological Data Sharing Service System (http://cdc.cma.gov.cn/home.do). Beijing station lies in the center of Beijing city. The BLH is from ERA-Interim by the European Centre for Medium-Range Weather Forecasts (ECMWF) (Persson and Grazzini, 2005). Daily BLH is the average of BLH values at 00:00, 03:00, 06:00, 09:00, 12:00, 15:00, 18:00, and 21:00 within one day.

3 Analyzing AERONET AOD

The AOD at 550nm is estimated using the spectral dependence of the AOD at the two nearest wavelengths, generally 500 and 675nm with the following equations (Ångström, 1964):

$$\tau(\lambda) = \beta\lambda^{-\alpha} \quad (1)$$

$$\alpha = \frac{-ln(\tau(\lambda_1)/\tau(\lambda_2))}{ln(\lambda_1/\lambda_2)} \quad (2)$$

$$\beta = \frac{\tau_\alpha(\lambda_1)}{{\lambda_1}^{-\alpha}} \quad (3)$$

where λ refers to the wavelength, $\tau(\lambda)$ represents the AOD at wavelength λ, and β the Ångström turbidity coefficient which equals the AOD at $\lambda = 1\mu m$, and α is the Ångström exponent (AE).

There are 2590 days with valid AOD data from the Beijing site of AERONET during the period 2001 to 2012, and we estimated the daily AOD at 550nm wavelength for these 2590 days using Eq. (1-3). The daily AOD at 550 nm wavelength is shown in Fig. 2. For the entire dataset the mean value is 0.66 ranging between 0.05 and 4.46. For the monthly data, both the mean and median of AOD values are highest in June, while both the maximum and minimum are highest in April. From April to August the AOD means exceed 0.7.

Beijing has a typical continental monsoon climate with four distinct seasons. Spring (March-May) experiences dust episodes from the Kumutage and Taklimakan deserts in western China and northerly winds from the Mongolian deserts (Sun et al., 2001). Summer (June-August) is characterized by relatively hot and humid weather with southerly winds. Autumn (September-November) is characterized by relatively clear weather. Winter (December-February) is dominated by cold, dry, and windy weather due to cold air advected by ~~from~~ the west Siberian anticyclone (Yu et al., 2013). High AOD values imply very high levels of air pollution and associated negative impacts on human health, while low AOD values represent good air quality. High AOD observed in spring (March-May) is

mainly due to dust events over Beijing (Cao et al., 2014). The highest AOD occurs in June despite the aerosol removal by monsoon precipitation, corroborating previous studies, e.g., Wang et al. (2010a) who showed that AOD is highest from June to August.

4 Estimating $PM_{2.5}$

4.1 Influence of the BLH and ambient RH

Based on the ground-based $PM_{2.5}$ observations from the U.S. embassy in Beijing from May 10, 2010, to December 6, 2011, a relationship with the observed AOD can be found. The relationship between AOD and $PM_{2.5}$ concentration has been investigated by many researchers. For example, Engel-Cox et al. (2004) developed simple empirical relationships between these two variables over the United States. The direct correlation between the moderate resolution imaging spectroradiometer (MODIS) AOD and $PM_{2.5}$ has been applied to estimate $PM_{2.5}$ across the global urban areas spread over 26 locations, and the results show that the relationship between $PM_{2.5}$ and AOD strongly depends on aerosol concentrations and ambient relative humidity (Gupta et al., 2006). Van Donkelaar et al. (2010) compared the original MODIS and multiangle imaging spectroradiometer (MISR) total-column AOD with ground-based measurements of daily mean $PM_{2.5}$, and both the MODIS and MISR instruments indicate some relationship between AOD and $PM_{2.5}$, both with spatial correlation coefficients R of 0.39. However, the AOD reflects aerosol optical extinction of the total column, while the $PM_{2.5}$ concentration measurements are at the surface. The correlation between AOD and $PM_{2.5}$ is strongly influenced by the vertical distribution of aerosols and the RH that impacts aerosol extinction coefficient. These two factors are related to atmospheric profiles, ambient conditions, as well as the size distributions and chemical compositions of aerosols, and they may have large spatial and temporal variations (Wang et al., 2010b). In order to reduce the uncertainties ~~caused by atmospheric profiles and ambient conditions~~, the atmospheric BLH and ambient RH have been introduced into the correlation analysis (Koelemeijer et al., 2006; Li et al., 2005; Liu et al., 2005; Wang et al., 2010b).

Under the assumption of a plane parallel atmosphere, AOD is the integral of the k_a at all altitudes along the vertical orientation, shown in Eq. (4). $k_a(\lambda, Z)$ represents the aerosol extinction coefficient at altitude Z and wavelength λ. In addition, assuming the vertical distribution of $k_a(\lambda, Z)$ as the negative exponent form is shown in Eq. (5). $k_{a,0}(\lambda)$ refers to the surface level aerosol extinction coefficient at wavelength λ, and H stands for the scale height of the aerosol. Substituting Eq. (5) to Eq. (4) we get Eq. (6). The $k_{a,0}(\lambda)$ could be calculated from AOD and H, and H could be approximately replaced by the atmospheric BLH. Therefore, the vertical correction, AOD/BLH, can reflect aerosol optical extinction at the surface level (Liu et al., 2005).

$$\tau(\lambda) = \int_0^\infty k_a(\lambda, Z)dz \quad (4)$$

$$k_a(\lambda, z) \approx k_{a,0}(\lambda)e^{\frac{-Z}{H}} \quad (5)$$

$$\tau(\lambda) \approx k_{a,0}(\lambda)\int_0^\infty e^{\frac{-Z}{H}}dz = k_{a,0}(\lambda) \times H \quad (6)$$

~~The correlation between aerosol extinction coefficient and $PM_{2.5}$ concentration is influenced by the chemical components of particles and RH of the ambient air.~~ Based on the previous studies (Im et al., 2001; Li et al., 2005; Wang et al., 2010b), the RH correction, $f(RH)$, can be represented as Eq. (7).

$$f(RH) = (1 - RH/100)^{-g} \quad (7)$$

Where g is an empirical fit coefficient, and it equals 1 in this study.

4.2 Correlation analysis

We compared the direct relationship between daily AERONET AOD at 550nm wavelength and $PM_{2.5}$ from May 10, 2010 to December 6, 2011, as shown in Fig. ~~1~~3(a). The correlation between the two datasets is rather poor with an R^2 of 0.28. After considering the influence of the BLH and ambient RH, the former obtained from the ECMWF assimilated analysis model (Persson and Grazzini, 2005) and the latter from the meteorological station, we find that the RH corrected $PM_{2.5}$ ($PM_{2.5}$ ×f(RH)) has a much higher correlation with the vertically corrected AOD (AOD/BLH), with an R^2 of 0.62, as shown in Fig. ~~1~~3(b). In addition, we compared the correlation coefficient (R^2) between AOD and $PM_{2.5}$ in this study to that established in some of the previous studies (Engle-Cox et al. (2004), Koelemeijer et al. (2006), Wang et al. (2010b), and Xin et al. (2014). It is found that our correlative model yields a higher correlation coefficient compared to these studies.

Based on the linear correlation in Fig. ~~1~~3(b), Eq. (8) coefficients were derived and Eq. (8) was then used to calculate the daily $PM_{2.5}$ in Beijing from 2001 to 2012 from the AERONET AOD. The results are shown in Fig. ~~2~~4(a), and the average and standard deviation for estimated $PM_{2.5}$ during these 12 years is 100.39 μg/m^3, and 55.67 μg/m^3, respectively. For the monthly data, the mean of $PM_{2.5}$ is highest in January and December, with the values of around 100~~6.44~~ μg/m^3. ~~The second highest mean of $PM_{2.5}$ occures in December with the value of 104.34 μg/m^3.~~ ~~Additionally, t~~The yearly averaged $PM_{2.5}$ from 2001 to 2012 is shown in Fig. 4(b), and it is highest in the year 2006, with the value of 111.4 μg/m^3. Clearly an increasing trend is present between 2001-2012, although no clear trends are detectable after the year 2004. With regard to the uncertainties of the estimated $PM_{2.5}$, we compared the estimated $PM_{2.5}$ to the ground-based $PM_{2.5}$ from the U.S. embassy in Beijing from May 10, 2010 to December 6, 2011. The average of the estimated $PM_{2.5}$ and ground-based $PM_{2.5}$ is 104 μg/m^3 and 93 μg/m^3 , respectively~~, and the correlation (R^2) between the two datasets is 0.33.~~. Han et al. (2007) investigated $PM_{2.5}$ concentrations in Beijing from 2001 to 2004 and found during summer of 2002, spring and autumn of 2003 that it was 79.6 μg/m^3, 111.6 μg/m^3 and 107.3 μg/m^3, respectively. In this study, the estimated $PM_{2.5}$ during summer of 2002, spring and autumn of 2003 was 73.7 μg/m^3, 99.9 μg/m^3, and 78 μg/m^3, respectively. Wang et al. (2009) found in summer and winter during 2005-2007 in Beijing that the average $PM_{2.5}$ was 102 μg/m^3. During the same period, our estimated $PM_{2.5}$ was 99 μg/m^3.

$$PM_{2.5} = (97569 \times AOD/BLH + 86.357)/f\,(RH) \quad (8)$$

5 Health effects

5.1 Beijing central area

The districts Chaoyang, Dongcheng and Xicheng in Beijing are adjacent and here collectively defined as the Beijing central area. The U.S. embassy is located in the Chaoyang district, and is also close to the Dongcheng and Xicheng districts. We have collected daily $PM_{2.5}$ data of 6 ground stations in these three districts from October 8, 2012, to November 13, 2012, from the study by Zhang et al. (2013). Fig. ~~3~~ 5 shows the daily $PM_{2.5}$ in the Dongsi and Tiantan stations in Dongcheng district, the Guanyuan and Wanshouxigong stations in Xicheng district, and the Aotizhongxin and Nongzhanguan stations in Chaoyang district. There is no obvious difference among the daily data in these six stations, and the high correlation between Dongsi station and other stations is shown in Fig. ~~4~~6. Therefore, we have used the daily $PM_{2.5}$ from the U.S. embassy station to represent the $PM_{2.5}$ concentration in the Beijing central area.

5.2 Concentration-response functions

Health effects of $PM_{2.5}$ have been derived from epidemiological cohort studies in a variety of geographical (principally urban) locations, mostly in the USA. Lelieveld et al. (2013) applied an epidemiological health impact function to calculate cardiopulmonary disease and lung cancer mortality attributable to air pollution in 2005. In the function, the concentrations of $PM_{2.5}$ are the yearly average in 2005, and the global population is also for the year 2005. Evans et al. (2012) used a concentration-response function for the association between $PM_{2.5}$ and mortality to calculate the lung cancer, cardiopulmonary disease, and ischemic heart disease mortality. Since we estimated $PM_{2.5}$ from 2001 to 2012 in the Beijing central area, we have calculated the yearly premature mortality caused by $PM_{2.5}$ using concentration-response functions that relate changes in pollutant concentrations to changes in mortality.

No epidemiologic study has estimated the association of long-term exposure to direct measurements of $PM_{2.5}$ with mortality from chronic cardiovascular and respiratory disease in Asia and other developing and emerging countries where annual average $PM_{2.5}$ exposures can exceed 100 μg/m^3 (Brauer et al., 2012). Previously, the functions for $PM_{2.5}$ have been based on the relationship between relative risk (RR) and concentrations defined by epidemiology studies where a log-linear (Ostro, 2004) and a linear model (Cohen et al., 2004) were used to calculate RR. However, the coefficients of these models are based on information from a single US cohort study – American Cancer Society Cancer Prevention II, with annual mean exposure levels below 22 μg/m^3. The form of the models used for global burden assessment was motivated largely by the concern that linear extrapolation would produce unrealistically large estimated of RR. ~~The Integrated Exposure-Response (IER) model that covers the global range of exposure is developed by integrating RR information from different combustion types that generate emissions of particulate matter, and it was able to take shapes similar to previous models such as linear and log-linear and a power function. In addition to these shapes, it also has the feature of flattening at high exposures. It was shown that it is a superior predictor especially for high $PM_{2.5}$ concentrations (Burnett et al., 2014).~~

5.3 Mortality estimation and discussion

Long-term exposure to $PM_{2.5}$ is associated with increased mortality from ischemic heart disease (IHD), cerebrovascular disease (stroke, CEV), chronic obstructive pulmonary disease (COPD), and lung cancer (LC), and increased incidence of acute lower respiratory infections (ALRI). Unfortunately, long-term cohort data from Beijing are not yet available. Therefore, we used the relationship between $PM_{2.5}$ and RR which are organized in bins from global burden of disease study 2010 (GBD 2010) (http://ghdx.healthdata.org/record/global-burden-disease-study-2010-gbd-2010-ambient-air-pollution-risk-model-1990-2010) ~~IER model~~ to calculate RR over the Beijing central area for causes of premature mortality in adults (>30 yrs): IHD, CEV, COPD and LC. In addition, the RR for ALRI was also calculated for infants (<5 yrs).

~~The IER model has the following form:~~

~~For $x < x_0, RR = 1$~~

~~For $x \geq x_0, RR = 1 + a \times \{1 - exp[-B \times (x - x_0)^p]\}$ (9)~~

~~where x is the exposure to $PM_{2.5}$ in μg/m^3, and x_0 is the threshold concentration below which it is assumed there is no additional risk. For very large x, RR~1+a. The power of $PM_{2.5}$, p, is used to predict risk over a very large range of concentrations. The parameters (a, B, p) are from the work of Burnett et al. (2014). Table 1 shows details of the IER model for estimating RR for five different diseases (IHD, CEV, COPD, LC, and ALRI).~~ We calculated the ~~value of x as the~~ yearly average $PM_{2.5}$ concentrations at

the U.S. embassy monitor station from 2001 to 2012, and it is shown in Fig. 4(b). ~~The threshold concentrations for different diseases are shown in Table 1.~~ Hence, we acquired the yearly RR of these five disease categories caused by $PM_{2.5}$. The fraction of the disease burden attributable to the risk factor (the attributable fraction), *AF*, is defined as (Anenberg et al., 2010; Ostro, 2004)

$$AF = (RR - 1)/RR \quad (~~10~~9)$$

—To calculate the number of premature mortality cases due to pollution $PM_{2.5}$, the *AF* is applied to the total number of deaths

$$\Delta Mort = y_0 \times Pop \times AF \quad (~~11~~10)$$

where $\Delta Mort$ is the change in annual mortality due to a pollutant (in our study $PM_{2.5}$). Pop is the total population with an age of >30 yrs or <5 yrs exposed to the pollutant. y_0 is the baseline mortality rate (BMR) for a given population and a specific disease. The household population and age distribution was obtained by the Beijing statistical yearbook for every year from 2001 to 2012 (see Fig. ~~5~~7), and the household registration record officially identifies a person as a resident of an area. Since 2010 there is an obvious increase of population in the Beijing central area, which does not really reflect the real population growth, but rather statistical data collection, because China carried out the fifth and sixth census in the year 2000 and 2010, respectively. The population record has been updated since the sixth census and therefore data before the year 2010 are not considered very accurate. Regarding BMR, we downloaded regional cause-specific mortality estimates (http://www.who.int/healthinfo/statistics/mortality_rawdata/en/index.html), and calculated them for IHD, CEV, COPD, LC, and ALRI in China. It should be emphasized that the calculations scale linearly with the BMR, so countries and regions with relatively high baseline mortality rates have proportionally higher mortality attributed to air pollution. As for the uncertainties of the mortality estimation, we mainly use the lower and upper bound of RR to calculate a minimum and maximum *AF*, and to further calculate a minimum and maximum mortality by Eq. (10).

Based on the health impact function, Eq. (~~11~~10), we have calculated the yearly premature mortality by IHD, CEV, COPD and LC for people >30 yrs, and ALRI for infants <5 yrs, as well as the corresponding uncertainties of the mortality estimation in the Beijing central area from 2001 to 2012, which is shown in Fig. ~~6~~8. The premature mortality due to CEV (>~~2580~~ 2225 deaths per year) is highest among the five diseases and the premature mortality by ~~COPD~~ IHD (>~~940~~ 788 deaths per year) is the second highest. In addition, the premature mortality by ALRI exceeds ~~35~~ 30 deaths per year. The annual premature mortality attributable to air pollution in the Beijing central area is shown in Fig. ~~7~~9, as well as the uncertainties of the estimation and the corresponding per capita mortality for all ages. The annual premature mortality is more than ~~4,900~~4,102 deaths/yr, and shows an increasing trend during 2001-2012, with the highest value of ~~7,783~~6,495 deaths in 2012. The average premature mortality attributable to $PM_{2.5}$ is around ~~6,100~~5,098 (2,463 - 6,621) deaths/yr for the period 2001-2012. The per capita mortality for all ages is ~~18~~15.0 per 10,000 person-year in 2012, higher than that of ~~16.6~~13.8 per 10,000 person-year in 2001. We calculate that the highest per capita mortality (~~18.6~~15.6 per 10,000 person-year) occurred in 2004. The per capita mortality for all ages due to $PM_{2.5}$ is around ~~18~~15.0 (7.3 -19.5) per 10,000 person-year for the period 2001-2012.. Since the population data are more accurate for the period 2010-2012, the sum of the premature mortality related to each disease and the corresponding ratio to the population section is shown in Table ~~2~~ 1 for the years 2010-2012.

It is found that the annual average premature mortality attributable to $PM_{2.5}$ is around ~~7,627~~6,382 (3,130 - 8,254) deaths/yr for the period 2010-2012 in the Beijing central area.. The per capita mortality under adults (>30 yrs) in the Beijing central area in 2010-2012 attributable to $PM_{2.5}$ by CEV (~~15.2~~13.1 per 10,000 person-year) is higher than any other disease. Further, the per capita mortality (for people >30 yrs and <5 yrs) attributable to $PM_{2.5}$ is around ~~27.6~~23.1 per 10,000 person-year for the period 2010-2012. The per capita mortality for all ages attributable to $PM_{2.5}$ by CEV (~~9.3~~8.0 per 10,000 person-year) is higher than by the other diseases as shown in Fig. ~~6~~8. For the period 2010-2012 the per

capita mortality for all ages due to $PM_{2.5}$ is around ~~17.9~~15.0 (7.4 -19.4) per 10,000 person-year. Although the average $PM_{2.5}$ from 2010 to 2012 (105.03 μg/m^3) is a little higher than during the period 2001 to 2012 (100.39 μg/m^3), the per capita mortality for all ages due to $PM_{2.5}$ for the period 2010-2012 is the same as that for the period 2001-2012. This is mainly because the per capita mortality is also influenced by the ratio between the population with an age of >30 yrs or <5 yrs and the population with all ages, and the ratio is 0.65 and 0.66 for the period 2010-2012, and 2001-2012, respectively.

~~There are few long-term cohort studies for chronic cardiovascular and respiratory disease and lung cancer in East and South Asia, where ambient exposures are often higher than in other parts of the world.~~ We have compared the estimation of the mortality due to $PM_{2.5}$ with past studies. Lelieveld et al. (2013) calculated megacity premature mortality due to air pollution, and found that the per capita mortality for all ages attributable to $PM_{2.5}$ is 11.79 per 10,000 person-year in Beijing. Anenberg et al. (2010) estimated the global premature mortality attributable to $PM_{2.5}$. According to the study of Anenberg et al. (2010), the per capita mortality for all ages attributable to $PM_{2.5}$ is about 10.8 per 10,000 person-year in Beijing. Since the study area addressed here is located in the Beijing central area with relatively high $PM_{2.5}$ concentrations, the per capita mortality (15.0 per 10,000 person-year) may be somewhat higher than in the entire city. If we assume that the range found in these studies, i.e., 11-15 per 10,000 person year, is representative for Beijing with a population of 20 million, it follows that the annual premature mortality rate due to $PM_{2.5}$ is approximately 22,000 - 30,000 per year. ~~shows is understandably~~ In addition ~~There are~~the few long-term cohort studies for chronic cardiovascular and respiratory disease and lung cancer in East and South Asia, show that ~~where~~ ambient exposures are often higher than in other parts of the world. ~~The IER model by Burnett et al. (2014) is very useful in this respect as it extends the risk estimates to higher exposures. While the IER model yields state-of-the-science predictions of the risk over a range of concentrations that prevail in China, some limitations of this approach remain. There are uncertainties due to lack of information on actual exposure to $PM_{2.5}$ for some source-specific RRs used to fit the model. Additionally, the IER model is developed for the entire global exposure to air pollution, not specific to Beijing, hence assuming that the toxicity of particulates is the same everywhere. Fortunately, the IER model considers the shape of the mortality RR functions at high ambient concentration, which is suitable for Beijing.~~ Therefore, it is a useful approach to estimate the mortality attributable to $PM_{2.5}$ by IHD, CEV, COPD, and LC, and ALRI in Beijing, showing that mortality due to long term exposure to air pollution is a severe problem and that air pollution abatement are needed urgently.~~.~~

6 Conclusion

We have analyzed the seasonal distribution and characteristics of AOD at 550nm wavelength in Beijing during the decadal period 2001-2012. Long-term $PM_{2.5}$ concentrations were calculated using an estimated linear relationship with AOD. The average and standard deviation of the estimated $PM_{2.5}$ from 2001 to 2012 is 100.39 μg/m^3, and 55.67 μg/m^3, respectively. Using concentration-response functions based on epidemiological cohort studies, we estimated the yearly mortality attributable to $PM_{2.5}$ by IHD, CEV, COPD and LC among people >30 yrs and that by ALRI among infants <5 yrs in the Beijing central area from 2001 to 2012. The estimated total mortality in central Beijing is ~~7,627~~ 6,382 deaths/year (average 2010-2012), and the per capita mortality for all ages is around ~~17.9~~15.0 (7.4 - 19.4) per 10,000 person-year. If we assume the range of our and previous studies to be representative of the city of Beijing, this implies a mortality attributable to $PM_{2.5}$ of about 22,000 - 30,000 persons per year. Considering~~per 10,000. Due to~~ the growing population ~~rate~~ of Beijing and the continued high levels of air pollution this study corroborates the urgency of air pollution abatement strategies ~~within its urban area~~.

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References

Akimoto, H.: Global air quality and pollution, Science, 302, 1716-1719, 2003.

Anenberg, S.C., Horowitz, L.W., Tong, D.Q., and West, J.J.: An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling, Environ. Health Perspect., 118, 1189-1195, 2010.

Ångström, A.: The parameters of atmospheric turbidity, Tellus, 16, 64-75, 1964.

Brauer, M., Amann, M., Burnett, R.T., Cohen, A., Dentener, F., Ezzati, M., Henderson, S.B., Krzyzanowski, M., Martin, R.V., Van Dingenen, R., van Donkelaar, A., and Thurston, G.D.: Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution. Environ. Sci. Technol., 46(2), 652-660, 2012.

~~Burnett, R. T., Pope, C. A., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., and Brauer, M.: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure, Environ. Health Perspect., http://dx.doi.org/10.1289/ehp.1307049, 2014.~~

Cao, L., Tian, W.Z., Ni, B.F., Zhang, Y.M., and Wang, P.S.: Preliminary study of airborne particulate matter in a Beijing sampling station by instrumental neutron activation analysis, Atmos. Environ., 36, 1951-1956, 2002.

Cao, C.X., Zheng, S., and Singh, R.P.: Characteristics of aerosol optical properties and meteorological parameters during three major dust events (2005–2010) over Beijing, China, Atmos. Res., 150, 129-142, 2014.

Cohen, A.J., Anderson, H.R., Ostro, B., Pandey, K.D., Krzyzanowski, M., Künzli, N., Gutschmidt, K., Pope III, C.A., Romieu, I., and Samet, J.M.: Urban air pollution, Comparative Quantification of Health Risks, 2, 1353-1433, 2004.

Crutzen, P.J.: Geology of mankind, Nature, 415, 23-23, 2002.

Dockery, D.W., Pope, C.A., Xu, X.P., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris Jr, B.G., and Speizer, F.E.: An association between air pollution and mortality in six US cities, New Engl. J. Med., 329, 1753-1759, 1993.

Engel-Cox, J.A., Holloman, C.H., Coutant, B.W., and Hoff, R.M.: Qualitative and quantitative evaluation of MODIS satellite sensor data for regional and urban scale air quality, Atmos. Environ., 38, 2495-2509, 2004.

Evans, J., van Donkelaar, A., Martin, R.V., Burnett, R., Rainham, D.G., Birkett, N.J., and Krewski, D.: Estimates of global mortality attributable to particulate air pollution using satellite imagery, Environ. Res., 120, 33-42, 2012.

Ezzati, M., Lopez, A.D., Rodgers, A., Vander Hoorn, S., and Murray, C.J.L.: Selected major risk factors and global and regional burden of disease, The Lancet, 360, 1347-1360, 2002.

Gupta, P., Christopher, S.A., Wang, J., Gehrig, R., Lee, Y., and Kumar, N.: Satellite remote sensing of particulate matter and air quality assessment over global cities, Atmos. Environ., 40, 5880-5892, 2006.

Han, L.H., Zhuang, G.S., Cheng, S.Y., and Li, J.: The mineral aerosol and its impact on urban pollution aerosols over Beijing, China, Atmos. Environ., 41, 7533-7546, 2007.

Han, X., Zhang, M., Gao, J., Wang, S., and Chai, F.: Modeling analysis of the seasonal characteristics of haze formation in Beijing, Atmos. Chem. Phys. Discussions, 13, 30575-30610, 2013.

HEI International Oversight Committee: Health effects of outdoor air pollution in developing countries of Asia: a literature review, Boston, MA, Health Effects Institute (Special Report No. 15), 2004.

Holben, B.N., Eck, T.F., Slutsker, I., Tanre, D., Buis, J.P., Setzer, A., Vermote, E., Reagan, J.A., Kaufman, Y.J., and Nakajima, T.: AERONET - A federated instrument network and data archive for aerosol characterization, Remote Sens. Environ., 66, 1-16, 1998.

Hu, M.G., Jia, L., Wang, J.F., and Pan, Y.P.: Spatial and temporal characteristics of particulate matter in Beijing, China using the Empirical Mode Decomposition method, Sci. Total Environ., 458, 70-80, 2013.

Im, J.S., Saxena, V.K., and Wenny, B.N.: An assessment of hygroscopic growth factors for aerosols in the surface boundary layer for computing direct radiative forcing, J. Geophys. Res.-Atmos., 106, 20213-20224, 2001.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., and Woollen, J.: The NCEP/NCAR 40-year reanalysis project, B. Am. Meteorol. Soc., 77, 437-471, 1996.

Kaufman, Y.J., Tanré, D., and Boucher, O.: A satellite view of aerosols in the climate system, Nature, 419, 215-223. 2002.

Koelemeijer, R.B.A., Homan, C.D., and Matthijsen, J.: Comparison of spatial and temporal variations of aerosol optical thickness and particulate matter over Europe, Atmos. Environ., 40, 5304-5315, 2006.

Li, C.C., Mao, J.T., Lau, A.K.H., Yuan, Z.B., Wang, M.H., and Liu, X.Y.: Application of MODIS satellite products to the air

pollution research in Beijing, Sci. China Ser. D, 48, Supp. II, 209-219, 2005.
Li, P., Xin, J.Y., Wang, Y.S., Wang, S.G., Li, G.X., Pan, X.C., Liu, Z.R., and Wang, L.L.: The acute effects of fine particles on respiratory mortality and morbidity in Beijing, 2004-2009, Environ. Sci. Pollut. R., 20, 6433-6444, 2013a.
Li, Z., Gu, X., Wang, L., Li, D., Xie, Y., Li, K., Dubovik, O., Schuster, G., Goloub, P., Zhang, Y., Li, L., Ma, Y., and Xu, H.: Aerosol physical and chemical properties retrieved from ground-based remote sensing measurements during heavy haze days in Beijing winter, Atmos. Chem. Phys., 13, 10171–10183, doi:10.5194/acp-13-10171-2013, 2013b.
Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K.: A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of Disease Study 2010, Lancet, 380, 2224-2260, 2012.
Liu, Y., Sarnat, J.A., Kilaru, V., Jacob, D.J., and Koutrakis, P.: Estimating ground-level $PM_{2.5}$ in the eastern United States using satellite remote sensing, Environ. Sci. Technol., 39, 3269-3278, 2005.
Lelieveld, J., Barlas, C., Giannadaki, D., and Pozzer, A.: Model calculated global, regional and megacity premature mortality due to air pollution, Atmos. Chem. Phys, 13, 7023-7037, 2013.
Ma, J.Z., Chen, Y., Wang, W., Yan, P., Liu, H.J., Yang, S.Y., Hu, Z.J., and Lelieveld, J.: Strong air pollution causes widespread haze-clouds over China, J. Geophys. Res., 115, D18204, 2010.
McDonnell, W.F., Nishino-Ishikawa, N., Petersen, F.F., Chen, L.H., and Abbey, D.E.: Relationships of mortality with the fine and coarse fractions of long-term ambient PM_{10} concentrations in nonsmokers, J. Expo. Anal. Env. Epid., 10, 427-436, 2000.
Ostro, B.: Outdoor air pollution, WHO Environmental Burden of Disease Series, 2004.
Persson, A., and Grazzini, F.: User Guide to ECMWF forecast products, Meteorological Bulletin, 3, 2005.
Pope III, C.A., Ezzati, M., and Dockery, D.W.: Fine-particulate air pollution and life expectancy in the United States, New Engl. J. Med., 360, 376-386, 2009.
Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Dentener, F., and Guibert, S.: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmos. Chem. Phys., 6, 5225-5246, 2006.
Shi, X.H., and Xu, X.D.: Progress in the study of regional impact of aerosol and related features of heavy fog in Beijing City (In Chinese), Chinese Journal of Geophysics, 55, 3230-3239, 2012.
Sun, J.M., Zhang, M.Y., and Liu, T.S.: Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960-1999: Relations to source area and climate, J. Geophys. Res., 106, 10325-10333, 2001.
Sun, Y.L., Wang, Z.F., Dong, H.B., Yang, T., Li, J., Pan, X.L., Chen, P., and Jayne, J.T.: Characterization of summer organic and inorganic aerosols in Beijing, China with an Aerosol Chemical Speciation Monitor, Atmos. Environ., 51, 250-259, 2012.
Sun, Y.L., Zhuang, G.S., Tang, A.H., Wang, Y., and An, Z.S.: Chemical characteristics of $PM_{2.5}$ and PM_{10} in haze-fog episodes in Beijing, Environ. Sci. Technol., 40, 3148-3155, 2006.
Sun, Y.L., Zhuang, G.S., Wang, Y., Han, L.H., Guo, J.H., Dan, M., Zhang, W.J., Wang, Z.F., and Hao, Z.P.: The air-borne particulate pollution in Beijing-concentration, composition, distribution and sources, Atmos. Environ., 38, 5991-6004, 2004.
vanDonkelaar, A., Martin, R.V., Brauer, M., Kahn, R., Levy, R., Verduzco, C., and Villeneuve, P.J.: Global estimates of ambient fine particulate matter concentrations from satellite-based aerosol optical depth: development and application, Environ. Health Perspect., 118, 847-855, 2010.
Wang, H.L., Zhou, Y.M., Zhuang, Y.H., Wang, X.K., and Hao, Z.P.: Characterization of $PM_{2.5}$/$PM_{2.5\text{-}10}$ and source tracking in the juncture belt between urban and rural areas of Beijing, Chinese Science Bulletin, 54, 2506-2515, 2009.
Wang, J., Yang, F.-M., Wang, D.-Y., and He, K.-B.: Characteristics and relationship of aerosol optical thickness and $PM_{2.5}$ concentration over Beijing (in Chinese), Journal of the Graduate School of the Chinese Academy of Sciences, 27, 10-16, 2010a.
Wang, J.F., Hu, M.G., Xu, C.D., Christakos, G., and Zhao, Y.: Estimation of citywide air pollution in Beijing, PloS one, 8, e53400. doi:10.1371/journal.pone.0053400, 2013.
Wang, Z.F., Chen, L.F., Tao, J.H., Zhang, Y., and Su, L.: Satellite-based estimation of regional particulate matter (PM) in Beijing using vertical-and-RH correcting method, Remote Sens. Environ., 114, 50-63, 2010b.
Winchester, J.W., and Mu-Tian, B.: Fine and coarse aerosol composition in an urban setting: A case study in Beijing, China, Atmos. Environ., 18, 1399-1409, 1984.
Xin, J.Y., Zhang, Q., Wang, L.L., Gong, C.S., Wang, Y.S., Liu, Z.R., and Gao, W.K.: The empirical relationship between the PM2.5 concentration and aerosol optical depth over the background of North China from 2009 to 2011. Atmos. Res., 138, 179-188, 2014.
Yang, S.J., Dong, J.Q., and Cheng, B.R.: Characteristics of air particulate matter and their sources in urban and rural area of Beijing, China, J. Environ. Sci., 12, 402-409, 2000.
Yu, X.N., Shi, C.Z., Ma, J., Zhu, B., Li, M., Wang, J., Yang, S.Y., and Kang, N.: Aerosol optical properties during firework, biomass burning and dust episodes in Beijing, Atmos. Environ., 81, 475-484, 2013.
Zhang, A., Qi, Q.W., Jiang, L.L., Zhou, F., and Wang, J.F.: Population Exposure to $PM_{2.5}$ in the Urban Area of Beijing, PloS one 8, e63486. doi:10.1371/journal.pone.0063486, 2013.

Zhang, J.Y., Zhang, Z., Meng, H.Y., Zhang, G.B., Zhao, W., and Pan, X.C.: Time-series analysis on relationship between air pollution and mortality from circulatory system diseases among registered residents in Chaoyang district of Beijing, Chinese journal of preventive medicine, 46, 139, 2012a.

Zhang, R., Jing, J., Tao, J., Hsu, S.-C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, doi:10.5194/acp-13-7053-2013, 2013.

Zhang, Y.J., Guo, Y.M., Li, G.X., Zhou, J., Jin, X.B., Wang, W.Y., and Pan, X.C.: The spatial characteristics of ambient particulate matter and daily mortality in the urban area of Beijing, China, Sci. Total Environ., 435, 14-20, 2012b.

Zhao, P.S., Dong, F., He, D., Zhao, X.J., Zhang, X.L., Zhang, W.Z., Yao, Q., and Liu, H.Y.: Characteristics of concentrations and chemical compositions for $PM_{2.5}$ in the region of Beijing, Tianjin, and Hebei, China, Atmos. Chem. Phys., 13, 4631-4644, 2013.

Zhao, X.J., Zhang, X.L., Xu, X.F., Xu, J., Meng, W., and Pu, W.W.: Seasonal and diurnal variations of ambient $PM_{2.5}$ concentration in urban and rural environments in Beijing, Atmos. Environ., 43, 2893-2900, 2009.

Zheng, S., Cao, C.X., and Singh, R.P.: Comparison of ground based indices (API and AQI) with satellite based aerosol products, Sci. Total Environ., 488, 398-412, 2014.

Zhu, L., Huang, X., Shi, H., Cai, X.H., and Song, Y.: Transport pathways and potential sources of PM_{10} in Beijing, Atmos. Environ., 45, 594-604, 2011.

~~Table 1. The IER model for estimating RR associated with long-term exposure to $PM_{2.5}$.~~

~~Disease~~	~~Age~~	~~Parameters~~			
~~Ischemic Heart Disease (IHD)~~	~~>30 yrs~~	~~a=1.65~~	~~B=0.0483~~	~~p=0.467~~	~~X_0=7.45~~
~~Cerebrovascular Disease (CEV)~~	~~>30 yrs~~	~~a=1.31~~	~~B=0.0120~~	~~p=1.274~~	~~X_0=7.36~~
~~Chronic Obstructive Pulmonary Disease (COPD)~~	~~>30 yrs~~	~~a=22.16~~	~~B=0.00110~~	~~p=0.697~~	~~X_0=7.34~~
~~Lung Cancer (LC)~~	~~>30 yrs~~	~~a=159.22~~	~~B=0.00020~~	~~p=0.759~~	~~X_0=7.35~~
~~Acute Lower Respiratory Infections (ALRI)~~	~~<5 yrs~~	~~a=2.38~~	~~B=0.00380~~	~~p=1.193~~	~~X_0=7.30~~

Table 1. Annual mortality attributable to air pollution by disease category, and the corresponding per capita mortality (IHD, CEV, COPD, and LC for people >30 yrs, and ALRI for infants <5 yrs) in 2010-2012 in the Beijing central area, as well as the uncertainty ranges of the total mortality estimation.~~.~~

Disease	IHD	CEV	COPD	LC	ALRI	Total	Uncertainty ranges of the total mortality	
							Minimum	Maximum
Annual mortality	1224	3426	1129	542	62	6382	3130	8254
Per capita mortality (per 10,000 person-yr)	4.7	13.1	4.3	2.1	3.9	23.1	11.3	29.8
Per capita mortality for all ages (per 10,000 person-yr)	2.9	8.0	2.7	1.3	0.1	15.0	7.4	19.4

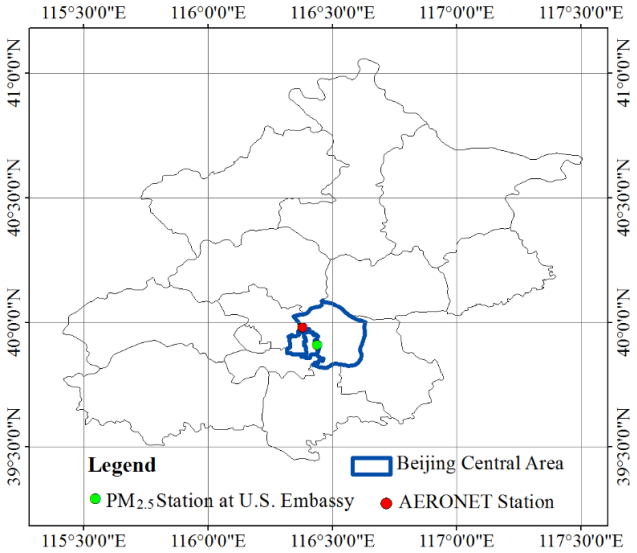


Fig. 1. Map of Beijing with AERONET and $PM_{2.5}$ stations. The green point refers to the $PM_{2.5}$ station at the U.S. Embassy, the red point to the AERONET station, and the blue line represents the Beijing central area, which includes the districts Chaoyang, Dongcheng and Xicheng.

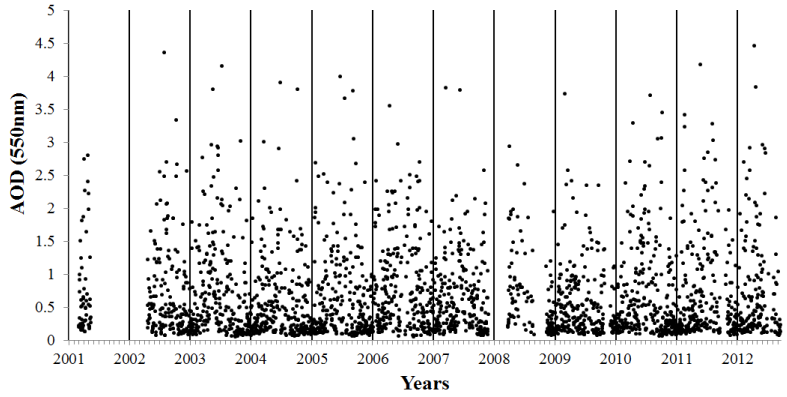
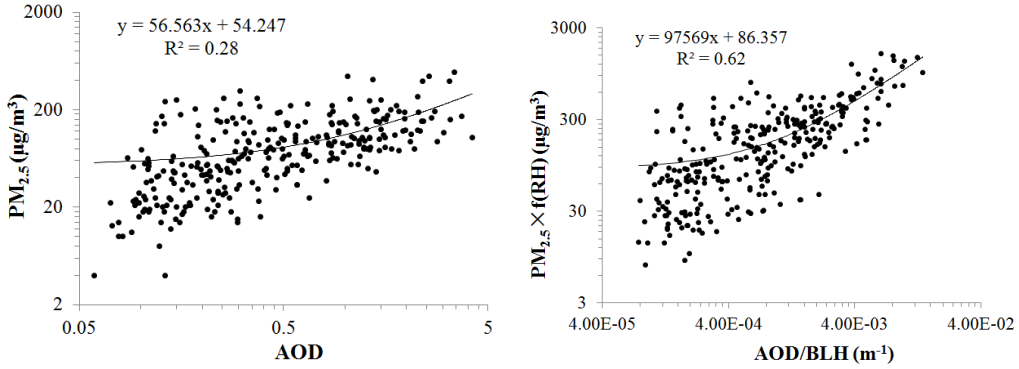
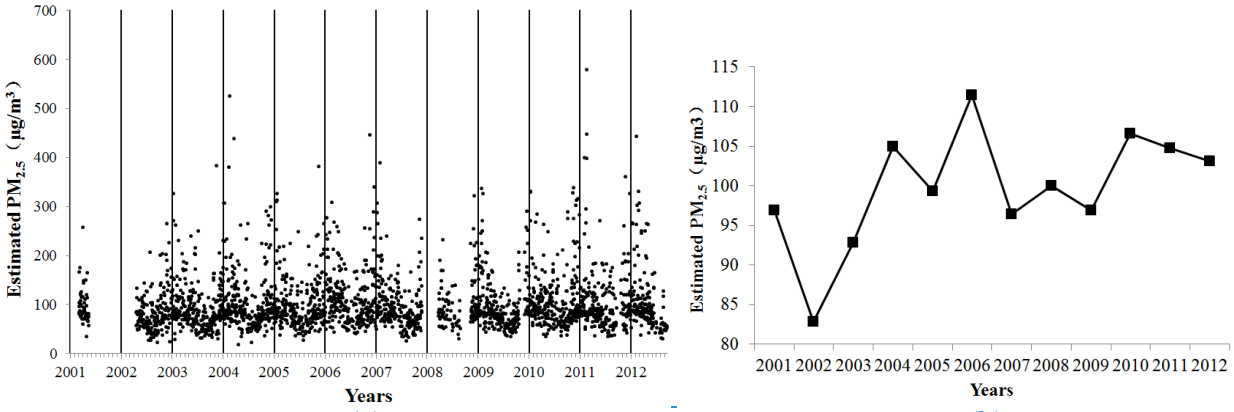


Fig. 2. Daily AOD at 550 nm wavelength from AERONET in Beijing from 2001 to 2012



(a) (b)

Fig. 13. Relationship between daily AERONET AOD and $PM_{2.5}$ from May 10, 2010 to December 6, 2011 in Beijing, (a) without BLH and RH correction, (b) with BLH and RH correction.



(a) (b)

Fig. 24. Estimated ~~daily~~ $PM_{2.5}$ from 2001 to 2012 in Beijing using AERONET AOD with BHL and RH correction~~.~~, (a) daily $PM_{2.5}$ (b) yearly averaged $PM_{2.5}$.

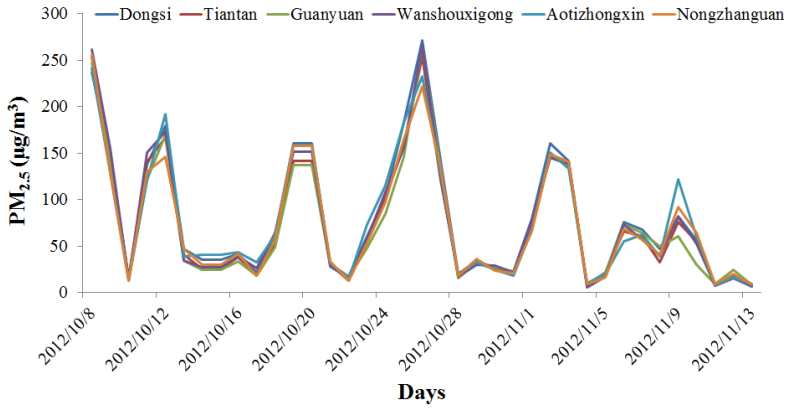


Fig. 35. Daily $PM_{2.5}$ from six ground stations in the Chaoyang, Dongcheng, and Xicheng districts.

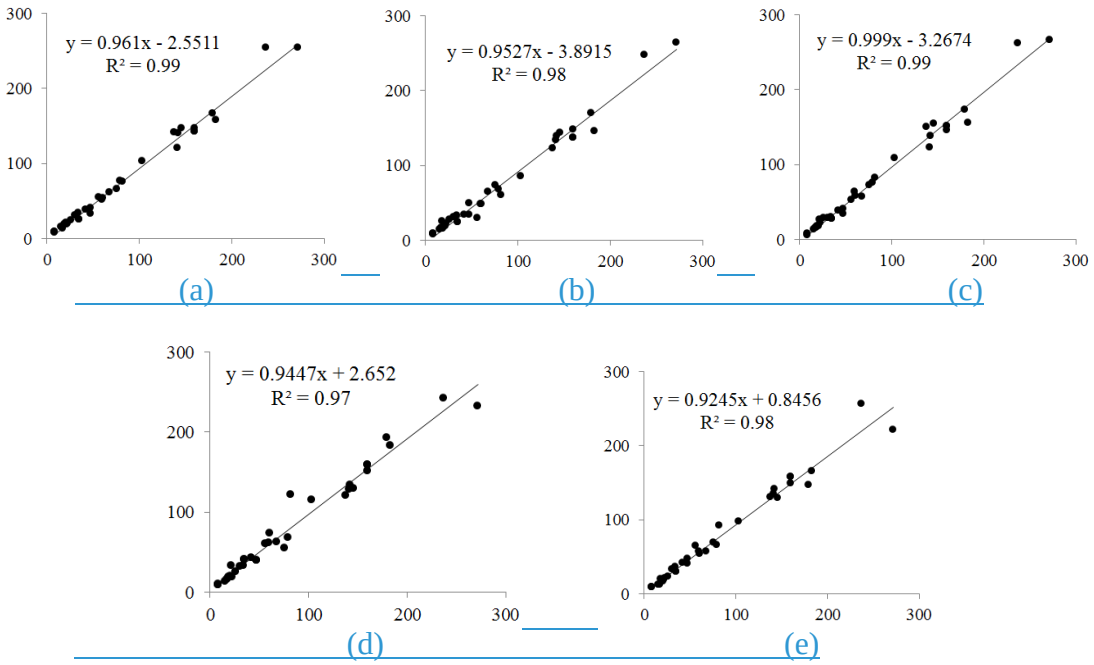


Fig. 46. Correlation of $PM_{2.5}$ between Dongsi station and the other five stations. (a) Dongsi and Tiantan, (b) Dongsi and Guanyuan, (c) Dongsi and Wanshouxigong, (d) Dongsi and Aotizhongxin, (e) Dongsi and Nongzhanguan.

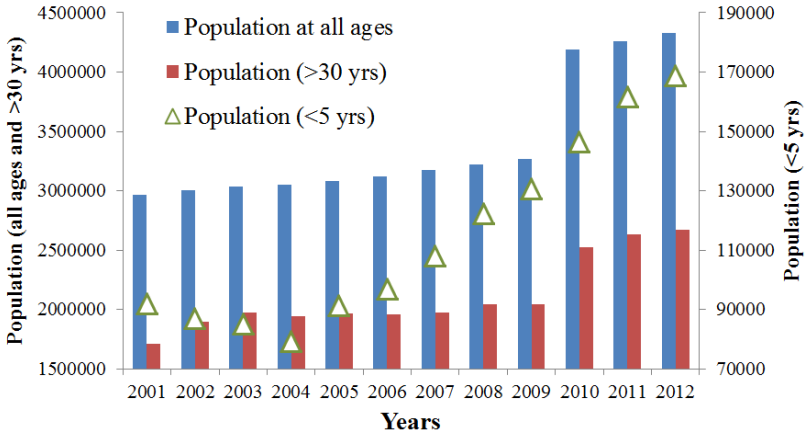


Fig. 57. Yearly registered population for all ages and of >30 yrs and <5 yrs in the Beijing central area.

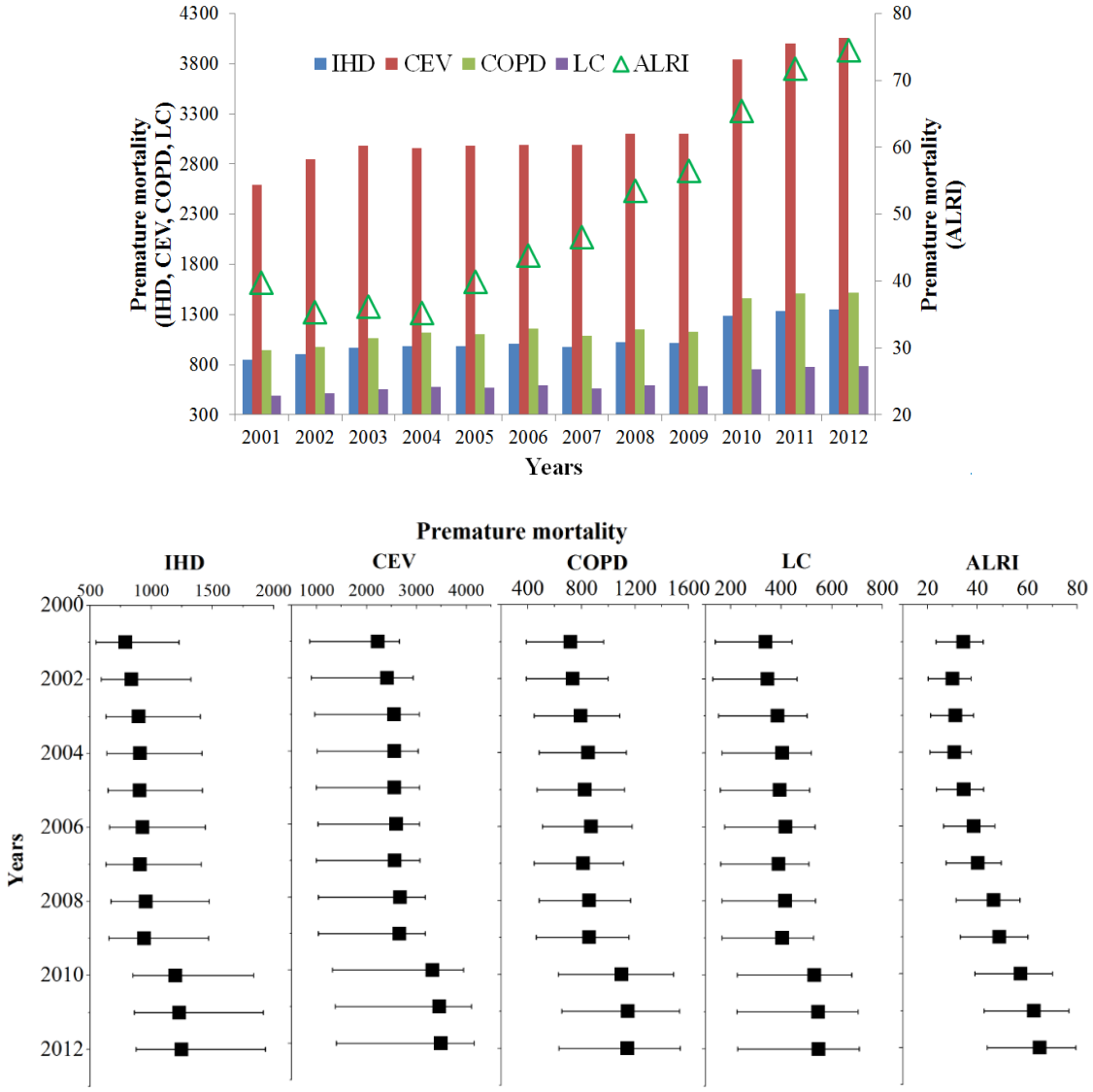
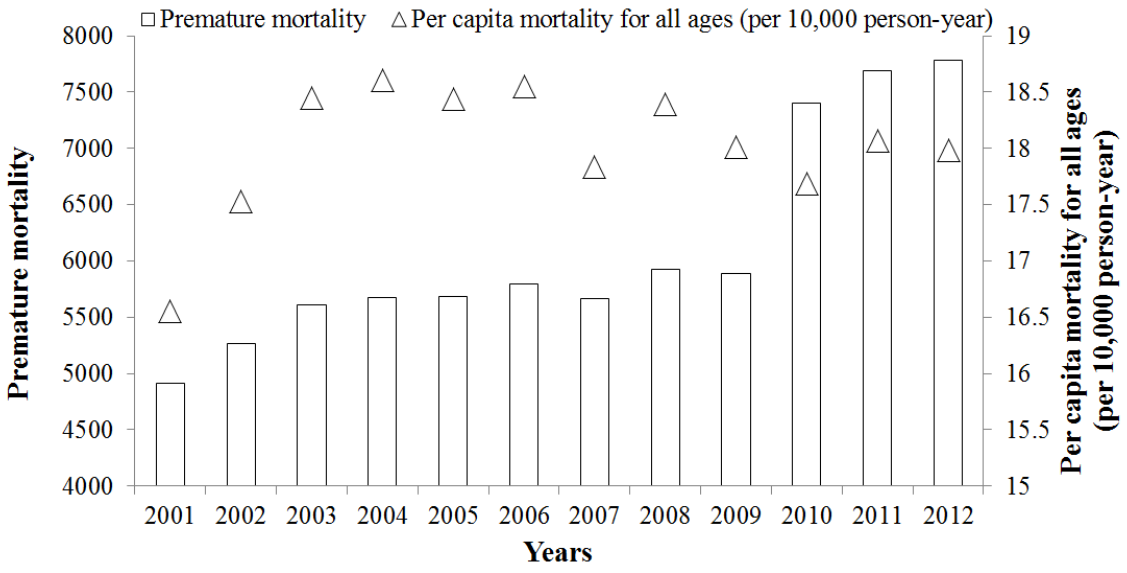


Fig. ~~6~~8. Yearly premature mortality attributable to IHD, CEV, COPD and LC for people >30 yrs, and ALRI for infants <5 yrs in the Beijing central area, as well as the corresponding uncertainties of the mortality estimation.



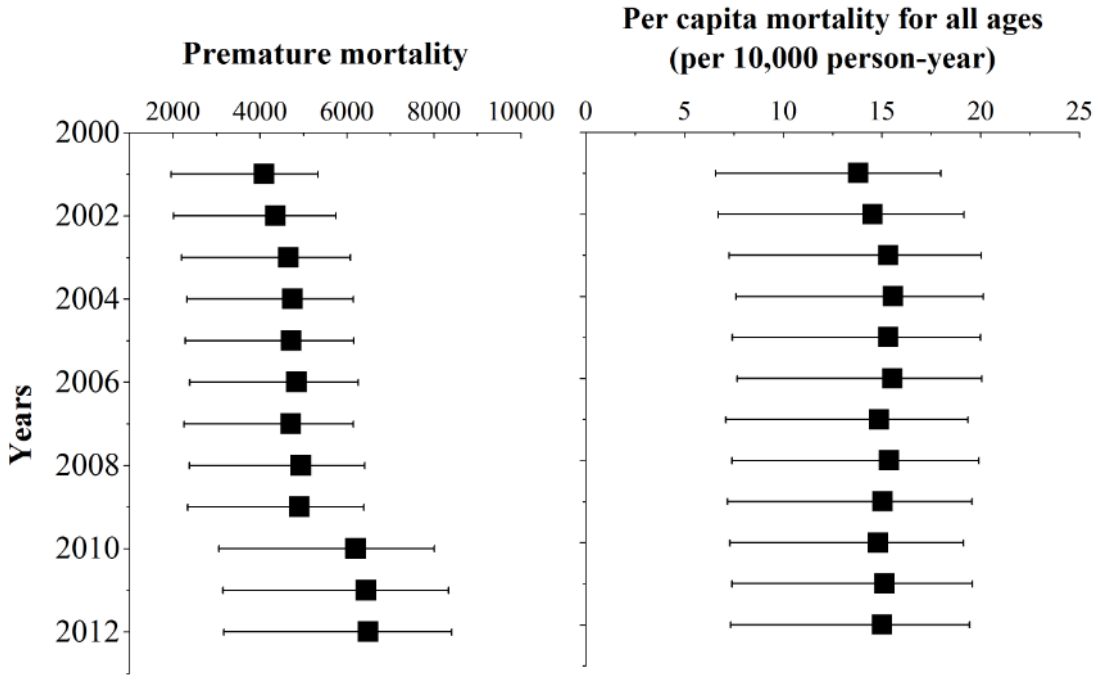


Fig. ~~7~~9. Annual premature mortality attributable to $PM_{2.5}$ and the ~~corresponding~~ per capita mortality for all ages for the period 2001-2012 in the Beijing central area, as well as the corresponding uncertainties.