All-Sky Direct Radiative Effects of Urban Aerosols in Beijing and Shanghai, China

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Abstract Aerosol particles can directly alter the radiation balance by scattering and absorbing incident solar radiation, thus decreasing the amount of light reaching the surface and increasing the fraction of diffuse radiation—the so-called 'aerosol direct radiative effect'. Using the Moderate Resolution Imaging Spectroradiometer aerosol products, the aerosol direct radiative effects under all-sky conditions in Beijing and Shanghai in 2007 were explored in this study. The total shortwave radiation was calculated using the Fu-Liou radiative transfer model, with the influence of clouds taken into account through sunshine-duration data, and the diffuse radiation was calculated with radiation decomposition models. Good correlation between measured and calculated total radiation was obtained at both cities, with an R greater than 0.9, and thus this calculation method was adopted to derive aerosol direct radiative effects. The presence of aerosols caused the mean

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total and diffuse solar radiation reaching the surface to change by −19.9% and +27.4% in Beijing, respectively, and by −18.4% and +6.5% in Shanghai. It was also found that, despite the strong negative correlation between aerosol optical depth and total radiation change, the diffuse radiation changes were determined predominantly by clouds. The effects of such changes induced by aerosols on plant productivity should be further studied.

Keywords: aerosol direct radiative effect, diffuse radiation, Fu-Liou radiative transfer model, sunshine duration

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1 Introduction

Aerosols are a complex chemical mixture of solid and liquid particles suspended in the atmosphere. They are currently renowned as one of the primary natural and anthropogenic climate forcing agents, inducing a remarkable effect on Earth’s climate (Forster et al., 2007). Aerosols change the radiation budget both directly, by scattering and absorbing incident solar radiation, and indirectly, by functioning as cloud condensation nuclei, thus changing clouds’ optical properties (Waggoner et al., 1981; Albrecht, 1989; Charlson et al., 1992; Schwartz, 1996). While exerting a negative radiative forcing on solar shortwave radiation, aerosols, through scattering in a clear atmosphere, can produce a larger fraction of diffuse radiation, which could lead to a higher gross photosynthetic rate due to the diffuse fertilization effect (Knohl and Baldocchi, 2008). All in all, aerosol radiative effects and their partitioning into diffuse and direct radiation play an important role in global environmental change and require extensive research.

As one of the largest developing countries, China has been experiencing rapid population expansion and economic growth, emitting large amounts of air pollutants into the atmosphere. Resulting from a combination of direct and precursor emissions and particular regional meteorological conditions, the concentrations of atmospheric aerosols are noticeably high in China (Tie and Cao, 2010; Qu et al., 2010; Cao et al., 2012), which has drawn substantial attention from scientists throughout the world. Aerosol direct effects on solar radiation over China have been extensively studied both by numerical simulations and observations (Xia et al., 2007; Wang et al., 2009; Li et al., 2010; Yan et al., 2010; Cai et al., 2011; Bai et al., 2011).

However, estimation of aerosol direct radiative effects under all-sky conditions still remains challenging because of the weak parameterizations of cloud processes in radiative transfer models and the lack of detailed observations for cloud optical properties (Quante, 2004; Nowak et al., 2008). There have been studies that have calculated all-sky radiation with a broadband radiation model by using sunshine duration data to parameterize cloud effects (Yang et al., 2006; Yang et al., 2007), which has proven to be both a convenient and pragmatic method. However, no such attempts have been made with complex multi-layer spectral models. In addition, previous studies have also identified the difficulties that radiative transfer models encounter in precisely modeling the diffuse radiation (Halthore et al., 2005; Randles et al., 2013). An alternative method is the adoption of the radiation decomposition model, which establishes the empirical relationship between diffuse radiation and clearness index and has proven to be effective. Following the pioneering work of Liu and Jordan (1960), large numbers of models have been developed (Jacovides et al., 2010) and adopted in studies estimating diffuse radiation (Gu et al., 2002; Matsui et al., 2008).

The objective of our study was to better understand the aerosol direct radiative effects under all-sky conditions in China. We adopted a complex radiative transfer model to calculate aerosol direct radiative effects in Beijing and Shanghai in 2007, with the influence of clouds taken into account through sunshine duration data, and the diffuse radiation was calculated with radiation decomposition models. To the best of our knowledge, the combination of sunshine-duration-calculated cloud effects and radiation decomposition models has not previously been reported.

2 Methods

2.1 Site description

Beijing and Shanghai, both with populations of more than 20 million, are the two largest cities in China. Beijing has a moderate continental climate with a dry and windy winter and a rainy summer, while Shanghai has a milder and wetter subtropical maritime monsoon climate. Over the past several decades, with the increasing metropolitan and industrial emissions of aerosol and precursor gases, both cities have suffered from severe air pollution; and aerosol particulates have been the major source of urban air pollution.

2.2 Methodology

2.2.1 Fu-Liou radiative transfer model

The Fu-Liou radiative transfer model (Fu and Liou, 1992, 1993; Rose and Charlock, 2002) calculates the spectral irradiance over the 15 shortwave bands from 175 mm to 4.0 μm and 12 longwave bands between 2200 and 10 cm^{-1} with a delta-four stream radiative transfer stream. Input parameters for Fu-Liou include aerosol optical depth (AOD), aerosol components, surface albedo, solar zenith angle, vertical profiles of ozone, and meteorological variables. Other important aerosol optical properties like single scattering albedo and asymmetry factor are calculated by the model based upon the input aerosol components. Other parameters, such as gas concentrations were set as the default value in the model and remained constant here.

2.2.2 Cloud effect

Defined by the World Meteorological Organization, sunshine duration refers to the length of time for which solar direct normal irradiance exceeds a threshold value of 120 W m^{-2}. In this work, the cloud effect on solar radiation was quantified using sunshine duration data to parameterize the shortwave band cloud transmittance τ_c as follows:

$$\tau_c = f(n / N), \qquad (1)$$

where n represents the actual sunshine duration, which is routinely measured at weather stations, and N is the potential sunshine duration, which we can derive from the Fu-Liou simulation, and f represents the function that is used to derive τ_c from n/N. The following formula, which was determined using observations of total radiation collected in Japan (Yang et al., 2007) and validated for photosynthetically active radiation estimation in China (Qin et al., 2012; Tang et al., 2013), was adopted here without modification:

$$\tau_c = 0.2495 + 1.1415(n / N) - 0.3910(n / N)^2. \quad (2)$$

Total radiation under all-sky conditions (total $\mathrm{SWR_{all}}$) was then calculated as follows:

$$\text{Total } \mathrm{SWR_{all}} = \tau_c \times \text{Total } \mathrm{SWR_{clear}}, \quad (3)$$

where $\mathrm{SWR_{clear}}$ and $\mathrm{SWR_{all}}$ refer to the modeled total radiation before and after we consider the cloud effects, respectively.

2.2.3 Diffuse radiation calculation

The Liu and Jordan type regression model (Liu and Jordan, 1960) relating the fraction of diffuse radiation in total radiation (K_d) and the clearness index (K_t), which refers to the ratio of total radiation to extraterrestrial radiation, was established here for the diffuse radiation calculation. Two types of regression models have mostly been developed for daily diffuse radiation (De Miguel et al., 2001). The first type is the piecewise empirical fits to the measured data. The second type is the equation put forward by Bartoli et al. (1982),

$$K_d = a + (1 - a) \exp[bK_t^c / (K_t - 1)], \quad (4)$$

where a, b, and c are all constants derived from regression.

The second type of regression model, which is much more concise, was chosen in our study to calculate diffuse radiation. The details of our model developing work is discussed in section 3.2.

2.3 Data

Three categories of data were adopted in this study. The first one was the aerosol properties. The most important aerosol optical property, AOD, was derived from both the MODerate-resolution Imaging Spectroradiometer (MODIS) Aqua and Terra products:

MOD08_L3 and MYD_L3. A gap-filling algorithm was adopted here to replace the missing value in the MODIS product (Garcia, 2010, 2011). We obtained the daily aerosol components from the Goddard Chemistry Aerosol Radiation and Transport model (Chin et al., 2009).

The second category was the other input data that the Fu-Liou radiative transfer model required. We derived vertical profiles of the vapor, pressure， and temperature from National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis data, and that of the ozone from the NCEP Stratospheric Monitoring Group Ozone Blended Analysis (SMOBA) product. Another important input variable, the surface albedo, was obtained from eight-day MCD43C3 products. We hypothesized that the surface albedo would remain unchanged within every eight-day period. The last category was the surface data routinely measured at China Meteorological Administration stations, including daily total radiation, daily diffuse radiation, and sunshine duration.

3 Results and analysis

3.1 Global radiation simulation

Using the methods and data described above, we calculated the daily global radiation in Beijing and Shanghai for the entire year of 2007 and compared these data with the measured values. The comparisons are shown in Fig. 1. The modeled values in Beijing (Fig. 1a) agreed well with the measured ones, with an R^2 of 0.9008 and an MBE (mean bias error: mean modeled value minus mean measured value) of only −0.97 W m^{-2}. In contrast, the modeled values in Shanghai (Fig. 1b) showed slightly weaker agreement with the measured ones, with an R^2 of 0.8134 and an MBE of −8.79 W m^{-2}. The reason for this difference may be related to the larger amount of clouds in Shanghai. The mean cloud transmittance in Beijing was about 0.732, which was much larger than that in Shanghai (0.597). This lower cloud transmittance may lead to larger uncertainty, and thus the weaker agreement between simulation and observation.

3.2 Regression model of diffuse SWR

Two radiation decomposition models were separately developed for Beijing and Shanghai based on the total radiation and diffuse radiation measured at each site. The model proposed by Bartoli et al. (1982) was chosen here for its conciseness. We adopted a MATLAB program to conduct the nonlinear regression analysis. The model developed for Beijing was

$$K_d = 0.1318 + 0.8682\exp[2.0596K_t^{2.6057} / (K_t - 1)], \quad (5)$$

and the model developed for Shanghai was

$$K_d = 0.2386 + 0.7614\exp[2.1353K_t^{2.6131} / (K_t - 1)]. \quad (6)$$

Figures 2a and 2b show the close correlation between K_d and K_t at the two cities. The equations and curves of the models we developed are also displayed. These two models were then adopted in Beijing and Shanghai, respectively, to calculate the diffuse radiation under different aerosol scenarios.

3.3 Sensitivity experiments

3.3.1 Aerosol radiative effects on radiation components

Aerosol direct radiative effects were estimated by conducting a sensitivity experiment in which we set the aerosol loading to be zero, i.e., AOD = 0, and calculated the radiation components without aerosols. The changes in radiation components after this experiment were then calculated and attributed to aerosol direct radiative effects. The following equation was adopted to define the relative changes, which was used to measure the extent of change for the daily radiation components:

$$\text{Radiation relative change} = (R_{\text{pre}} - R_{\text{no}}) / R_{\text{no}}, \tag{7}$$

where R_{pre} is the daily total/diffuse radiation with present aerosol loading, and R_{no} is the daily total/diffuse radiation component without aerosols.

Figure 3 exhibits the aerosol induced monthly relative changes of total and diffuse radiation in Beijing (Fig. 3a) and Shanghai (Fig. 3b). We found that aerosols decreased total radiation on all occasions but increased diffuse radiation in most cases. This is consistent with the mechanism of aerosol direct radiative effects, in that aerosols scatter and absorb solar radiation and thus cause it to drop and, meanwhile, increase the proportions of diffuse radiation through scattering. The reduction in annual mean total radiation was 37.4 W m^{-2} in Beijing and 30.7 W m^{-2} in Shanghai. Furthermore, aerosols increased the annual mean diffuse radiation by 14.8 W m^{-2} in Beijing and 5.5 W m^{-2} in Shanghai. The relative changes of total and diffuse radiation were −19.9% and 27.4% in Beijing, respectively, and −18.4% and 6.5% in Shanghai. It can be seen that aerosol direct radiative effects were stronger in Beijing, particularly on diffuse radiation, which may be attributed to the higher cloud transmittance in Beijing, and is discussed in the following section.

3.3.2 Correlation between τ_c, AOD, and radiation relative changes

The correlation between τ_c, AOD, and radiation relative changes is illustrated in Fig. 4. It shows that AOD and τ_c played different roles in determining the radiation relative changes. Total radiation, as expected, decreased significantly with the increased AOD, and those two variables had a strong correlation with an R of about 0.745. In contrast, τ_c had no relationships with total radiation relative changes (Fig. 4b). On the contrary, τ_c was dominant in determining diffuse radiation, since large diffuse radiation relative changes only occurred on days with high τ_c and the correlation between AOD and diffuse radiation relative changes was noticeably weak (Figs. 4c and 4d). The reason behind this phenomenon may be that, on days with low τ_c, clouds not only significantly decreased the solar radiation that reached the surface, but they also caused a high K_d. In this case, even though the presence of aerosols increased K_d, or even barely did because K_d had already approached unity on days with very low τ_c, the combination of the large decrease of the total radiation (AOD and total radiation were negatively correlated, as discussed above) and the small increase in K_d led to the insignificant increases or even reduction of diffuse radiation.

It is worth noting that the diffuse relative change reached its peak around the τ_c value of 0.93. Then, it tended to drop with increased τ_c (Fig. 4d). This was caused by the radiation decomposition models we developed and adopted (Eqs. (5) and (6)). It can be seen from Fig. 2 that, as k_t approached 0, the absolute values of the slopes of both models' curves approached 0 as well, meaning that, in this region, K_d remained relatively steady with the changes of K_t. Thus, on days with very high τ_c and K_t, the presence of aerosols did not result in apparent increases in K_d like it did on days with smaller K_t. It can be concluded that, despite the fact that clouds mostly inhibited the positive radiative effects of aerosol on diffuse radiation, the positive effect was strongest on days with a small amount of cloud.

4 Conclusion

The direct radiative effects of urban aerosols in Beijing and Shanghai in 2007 were quantitatively estimated in this study using the Fu-Liou radiative transfer model, with the influence of clouds taken into account through sunshine-duration data, and the diffuse radiation was calculated with diffuse radiation empirical models. To the best of our knowledge, this method has not previously been reported and, by adopting it, we were able to demonstrate how clouds influenced the aerosol direct radiative effect.

It was found that aerosols decreased total radiation on all occasions but increased diffuse radiation in most cases. This radiative effect not only showed great temporal variability, but also varied significantly in the two cities we studied. The relative changes of annual total and diffuse radiation were −19.9% and 40.3% in Beijing, respectively, and −18.4% and 9.0% in Shanghai. AOD was found to have a strong negative correlation with the relative change of total radiation, but a considerably weak correlation with that of diffuse radiation. Cloud transmittance, on the other hand, played a dominant role in determining diffuse radiation relative changes. On days with very low cloud transmittance, aerosols not only barely increased the diffuse radiation, but even caused it to drop. Furthermore, it was also found that the strongest aerosol positive radiative effect on diffuse radiation occurred on days with a small amount of cloud. Such radiation changes could exert an influence on plant productivity and, thus, on the global carbon cycle, which should be further studied.

Uncertainties may arise from data limitation and model simplification. Firstly, without using specific cloud data, our modeling results could not present the radiation variation on cloudy days, but merely provide a relatively precise daily mean value. Secondly, we derived daily AOD from instantaneous MODIS aerosol observations and, furthermore, a gap-filling algorithm was adopted to substitute the missing value in the MODIS product. These could all have led to certain inaccuracies. Thirdly, the adoption of the formula developed by Yang et al. (2007) without any modification may also have resulted in uncertainties in radiation modeling. Finally, partitioning total radiation into direct and diffuse components with empirical models, instead of basing it on the radiation transfer mechanism, would also have brought some uncertainties.

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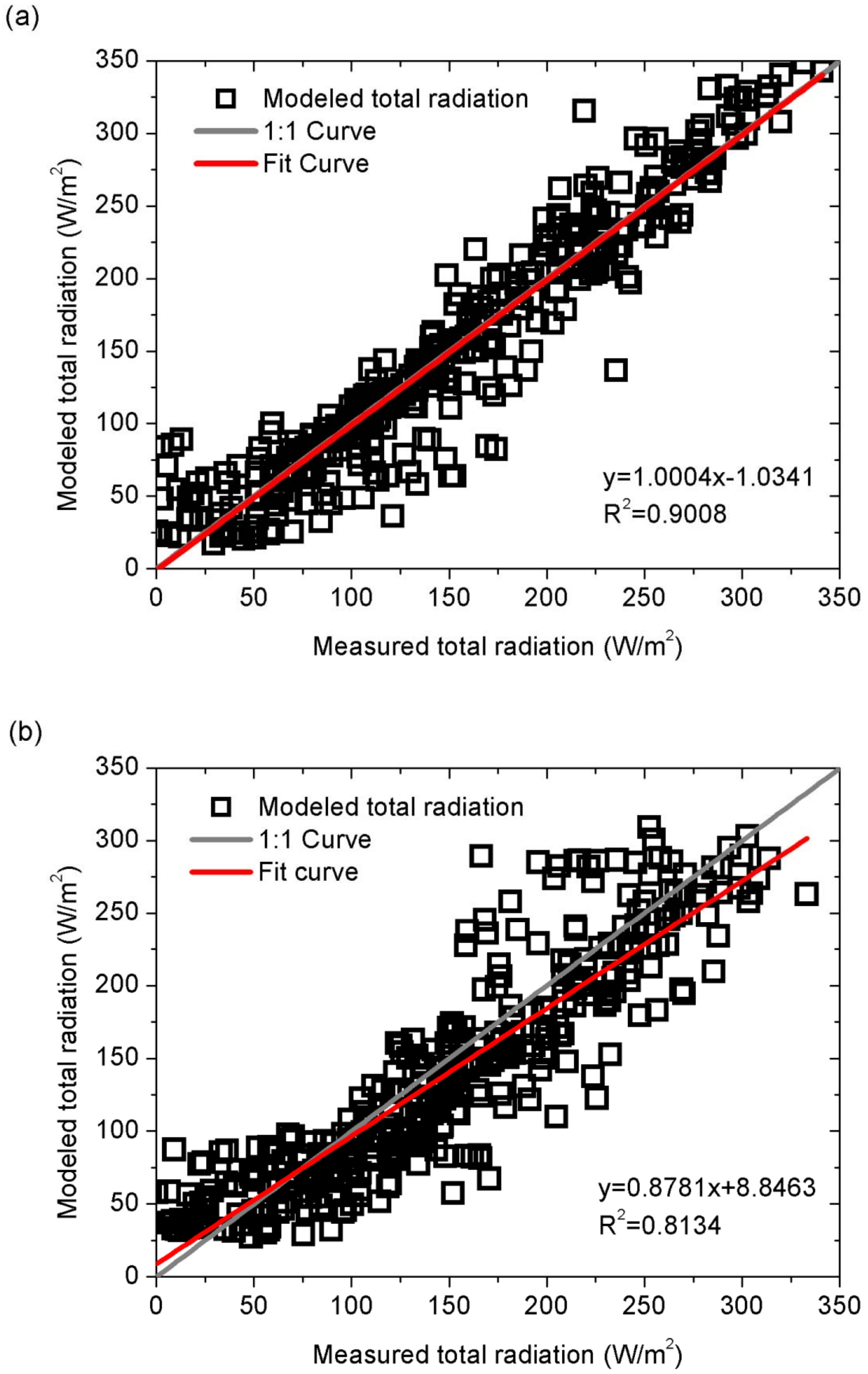

Figure 1 Scatter plots of the modeled daily total radiation versus the measured daily total radiation in 2007 at the study sites: (a) Beijing; (b) Shanghai.

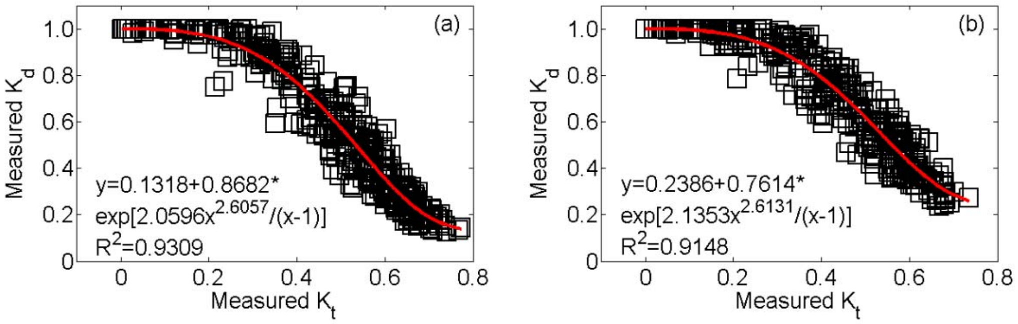

Figure 2 (a) Scatter plot of measured clearness index versus measured diffuse fraction in Beijing; (b) Scatter plot of measured clearness index versus measured diffuse fraction in Shanghai. Note that red curves are the plots of our newly developed diffuse radiation empirical models.

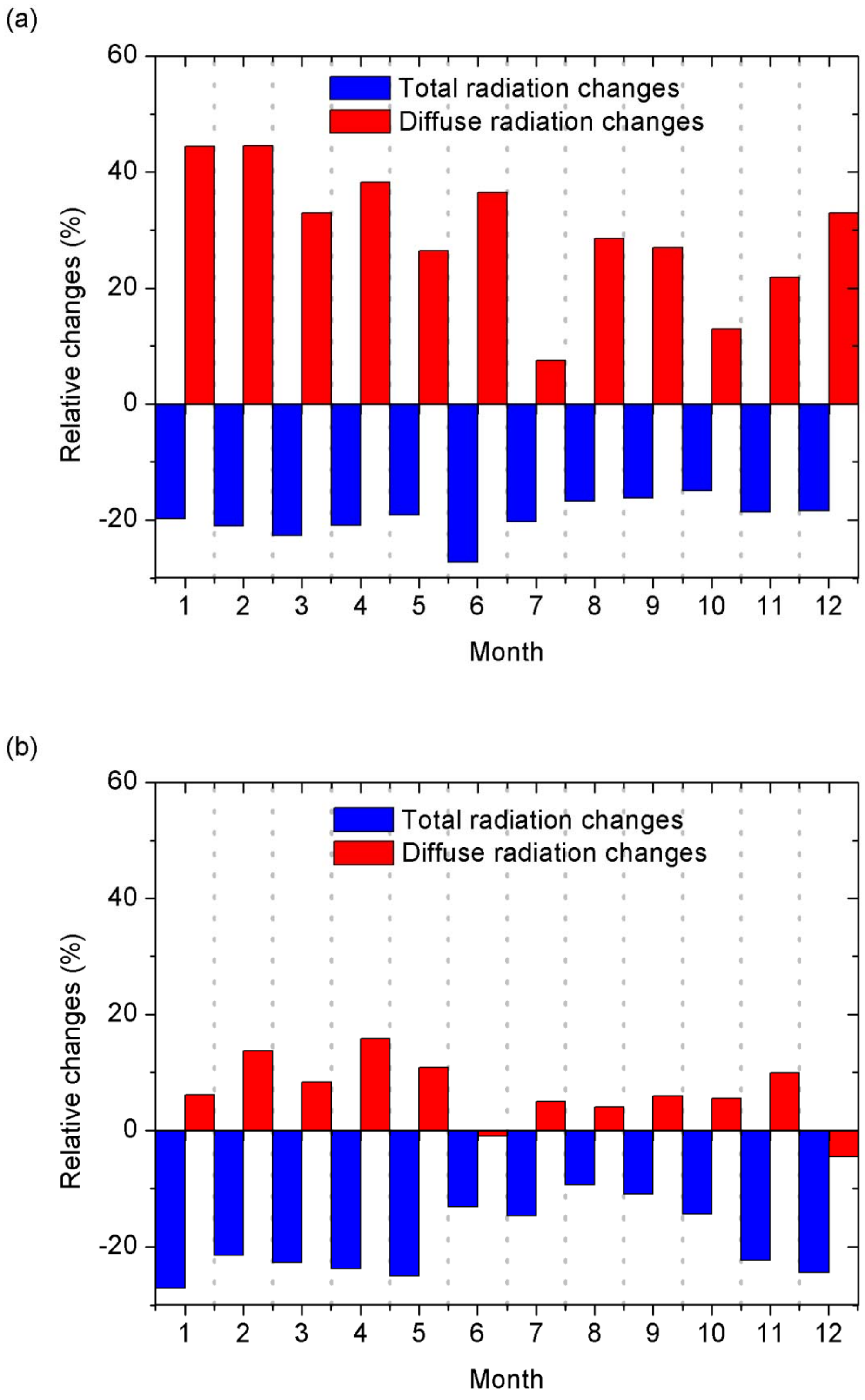

Figure 3 Aerosol-caused relative changes of daily radiation components at the study sites: (a) Beijing; (b) Shanghai.

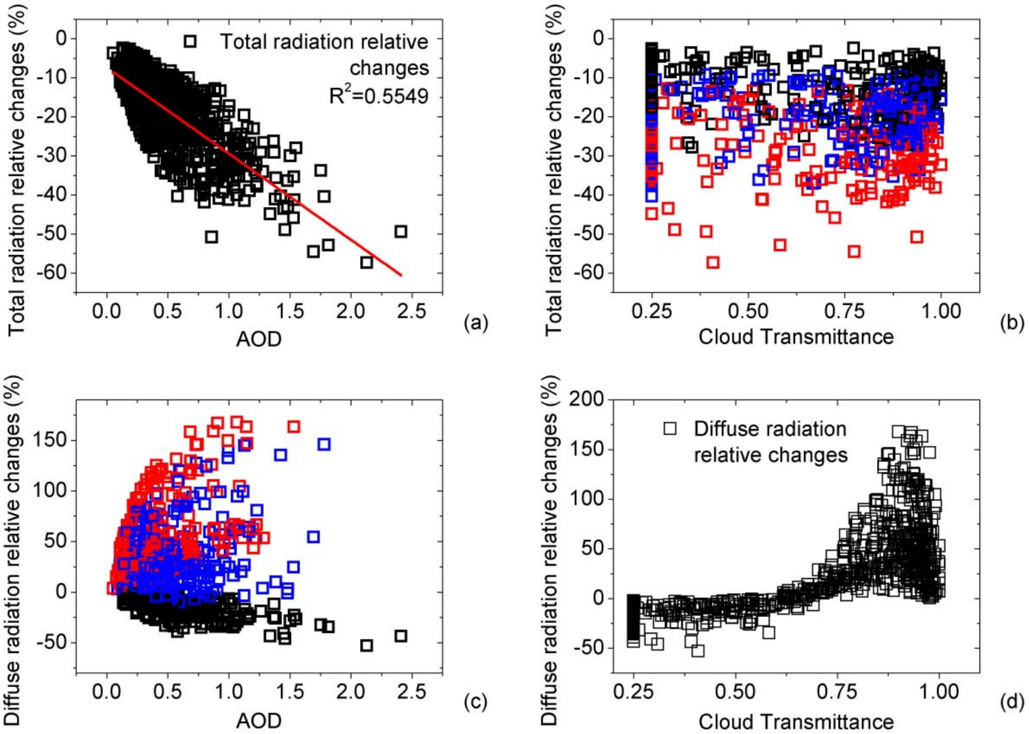

Figure 4 (a) Scatter plot of Aerosol Optical Depth (AOD) versus total radiation relative changes; (b) Scatter plot of cloud transmittance versus total radiation relative changes. Note that red squares represent days with AOD larger than 0.7; blue squares, days with AOD between 0.4 and 0.7; and black squares, days with AOD smaller than 0.4. (c) Scatter plot of AOD versus diffuse radiation relative changes. Note that red squares represent days with cloud transmittance larger than 0.9; blue squares, days with cloud transmittance between 0.6 and 0.9; and black squares, days with cloud transmittance smaller than 0.6. (d) Scatter plot of cloud transmittance versus diffuse radiation relative changes.