SUPPLEMENTARY MATERIAL

Correlation analysis between AOD and cloud parameters to study their relationship over China using MODIS data (2003-2013): Impact on cloud formation and climate change

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S1. Literature review

Alam *et al.* (2010) analyzed the effect of aerosols on clouds and showed enhanced formation of clouds was due to both smoke and pollution over Karachi and Lahore in Pakistan. Balakrishnaiah *et al.* (2012) noticed high AOD during the summer-monsoon seasons over major cities in southern India and studied the relationship to various cloud parameters. Yi *et al.* (2012) found an increasing variation in cloud fraction (CF) with the increase of AOD over ocean regions under observation, while the reverse result was noticed in the model simulation. Hoeve *et al.* (2012) investigated an increase in COT with increasing AOD at low AODs, and a decrease in COT with increasing AOD at higher AODs. This increase was attributed to a combination of microphysical and dynamical effects, whereas, the decrease was due to the dominance of radiative effects that thin and darken clouds.

Kumar (2013) also analyzed that AOD and CF correlation increases for those regions which have more particulate pollution due to dust, biomass, industrial and domestic activities over the northeast areas of India. Another study by Kumar (2014) found that AOD showed positive correlation with CF and COD and negative correlation with cloud top temperature (CTT) and cloud top pressure (CTP) over an urban region of northern India (Delhi) for the period 2003–2012. Alam *et al.* (2014) have recently studied the relationship between AOD and cloud properties in different locations of Pakistan. They noticed an increasing trend in CF with AOD over urban regions during the period of study from 2001 to 2011 as well as investigated the impact of aerosols on warm and cold clouds. Also very recently, Tang et al. (2014) showed positive correlation between AOD and cloud effective radius (CER) over open oceanic regions of East China, suggesting that the influence of background weather conditions and general circulations need to be considered when studying the interactions between aerosol and cloud.

S2. Methodology

The Ångström exponent (AE) is a qualitative indicator of aerosol particle size (Kaufman and Nakajima, 1993). The value of AE less than 1 indicates particle size dominated by coarse-mode aerosols with radii larger than 0.5 μm, which are usually associated with dust and sea salt, and values larger than 2 indicates particle size dominated by fine-mode aerosols with radii less than 0.5 μm that are usually associated with urban pollution and biomass burning (Eck *et al.,* 1999). AE from the MODIS product can be employed for qualitative judgment of the aerosol mode (Ångström, 1961). Assuming the size distribution of aerosol particles fits Junge-distribution, the relationship between AOD and AE can be expressed as:

$$\tau_a(\lambda) = \beta\lambda^{-\alpha} \qquad (1)$$

where $\tau_a(\lambda)$ is the AOD at λ, β is Ångström turbidity and α is Ångström wavelength exponent inversely related to the effective radius of aerosol particles. MODIS AOD at 550 nm can be obtained by interpolation between AODs at 470 nm and 660 nm based on the above formula.

S3. Relationship between AOD and water vapor (WV)

The behavior of aerosols in response to changes in WV was investigated and it provided an opportunity to understand the impact of aerosols on Earth's atmosphere (Myhre *et al.,* 2007). The time series plot for AOD and WV is shown in Fig. S1 for all the selected regions in China during the period 2003–2013 represents increasing and decreasing pattern simultaneously. This is in accordance with the findings of Ranjan *et al.* (2007), Balakrishnaiah *et al.* (2012), and Kumar *et al.* (2009) reported over Indian subcontinent and Alam *et al.* (2010, 2014) for Pakistan. The direct effect results in radiation scattering due to an increase in aerosol particle-size, accompanied by the uptake of WV. Changes in water uptake of aerosols can therefore, lead to changes in both direct and indirect radiative forcing on climate (IPCC, 2007). El-Askary and Kafatos (2008) have found that aerosols cause a reduction in cloud droplet size and hence lead to suppression in precipitation.

The statistical regression data obtained from the correlation analysis between AOD and WV were shown in Table 3. It clearly demonstrates from Table 3 that the calculated coefficient of determination (R^2) values clearly shows the positive correlations for the above relation. The highest positive R^2 value was found in the increasing order for the regions BJ (0.436), KM (0.372) and HH (0.185) with its lowest value in the decreasing order for the regions CD (0.008), XN (0.006) and NN (0.005). Similarly, the correlation coefficient (r) values were noticed to be positive for all the regions, except for the regions XN, LH and NN where r value found to be negative. The highest positive correlation coefficient value was noticed to be 0.66 and 0.61 for the regions BJ and KM, respectively followed by HH (+0.43) and NJ (+0.31) and maximum negative r value over region LH (-0.27). The maximum slope of the trendline for the all the regions computed was found to be 0.127 and 0.101 between AOD and WV observed for the regions BJ and KM, respectively were statistically significant at 95% confidence level ($p<0.05$). The student's t-Test probability was performed to the data and the maximum (minimum) probability $|t|$ was found to be 6.0E-18 (1.4E-49) for NJ (GZ). This uncertainty may occur in the AOD due to contamination by both low and high altitude cirrus clouds. Lee *et al.* (2009) concluded that cirrus clouds can increase the uncertainty in measured AOD by up to 20%.

The water absorbing ability of aerosols (hygroscopic nature) depends upon the particular mixing of different types of aerosols particles as well as on the meteorological conditions (Kaufman *et al.,* 2005; Kaufman and Koren, 2006; Aloysius *et al.,* 2009). Lee *et al.* (2009) suggested that water uptake of atmospheric aerosols can alter both the size and composition of particles and hence their optical particles. Wright *et al.* (2010) and Xie *et al.* (2011) depicted that the radiative forcing on climate significantly influenced by the changes in water uptake behavior of aerosols and hence cloud formation. Guo *et al.* (2014) and Luo *et al.* (2013) concluded that natural and anthropogenic aerosols over China play an important role in influencing the convective cloud formation and hence causing climatic implications to the overall hydrological cycle.

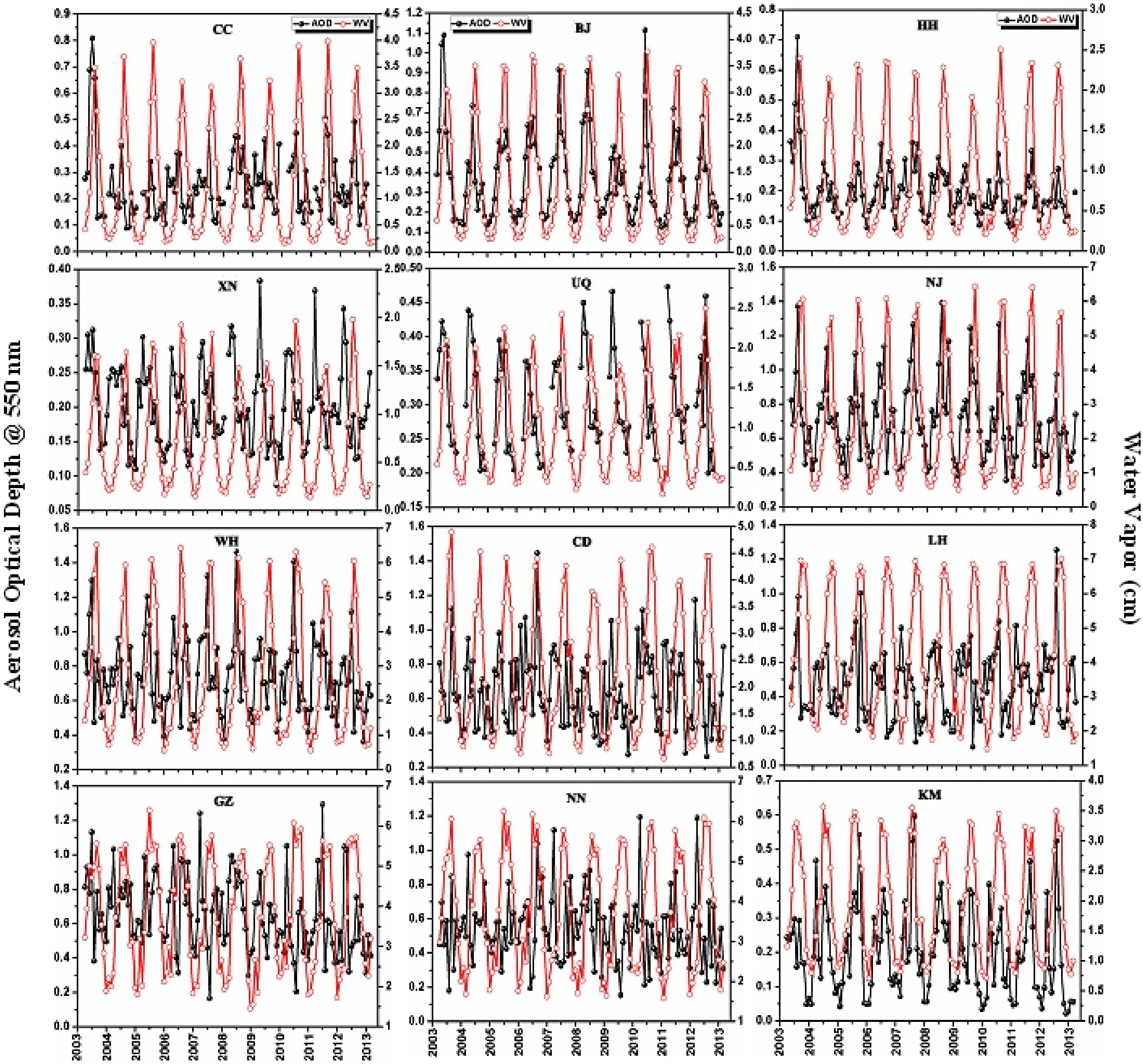

Fig. S1. Relationship between AOD_{550} and water vapor for the 12 selected sites in China during the period of 2003–2013.

S4. Relationship between AOD and CTP

The time series variation between AOD and CTP shown in Fig. S2 also represents the similar variation with increase in AOD there was a significant decrease in the CTP value over the selected regions of China. It is clear from Fig. S2 that CTP reaches a maximum value during January with a minimum value in July, where the AOD pattern is exactly opposite to that of CTP for most of the regions (see Table1). The correlation coefficient between AOD and CTP for various regions in China over the period of study is shown in Table 3. The R^2 value noticed to be maximum for region of BJ (0.462) and minimum over CD (0.008). Out of 12 selected regions in China, a strong negative correlation coefficient was found over 9 regions of which the maximum negative r value obtained for the region B (-0.68) followed by the regions NJ (-0.53), KM (-0.53), and HH (-0.51). Earlier researchers have reported that except for some regions of low AOD, CTP decreased in most of the areas (higher cloud altitude) as AOD increased (Alam *et al.,* 2014 and references therein). This might have resulted from the suppression of the precipitation by increasing cloud lifetime and thus also affecting the cloud albedo and changing the CTP (Ali *et al.,* 2013). Further, Ramanathan *et al.* (2001) and Lee *et al.* (2009) suggested since the CER increases with decrease of CTP and thereby, decreasing the CTP with increasing AOD.

A very low slope of the decreasing trend was found in most of the regions indicates the trend is almost a straight line and the maximum (minimum) slope of the decreasing trendline was -0.001 (-0.0008) noticed for the regions BJ and NJ (XN) which is 95% significant level. The probability value was found to be low (high) for XN (LH) which is 1.6E-99 (1.6E-64) during the period of study. Alam *et al.* (2010) investigated that at lower latitudes, there was a significant decrease in CTP in relation to AOD (i.e., negative correlation), and while at mid latitudes this decrease was only moderate. Our analysis indicated a strong negative correlation for AOD with CTP in the southern regions and a moderate positive correlation in the northern regions of China which is in agreement with the previous findings reported for south Asia. This co-variation of AOD with CTP may be attributed to large-scale meteorological variations. Koren *et al.* (2005) and Tripathi *et al.* (2007) reported the relation between vertical wind velocity and CTP. They suggested that the relationship is less influenced by meteorology during winter due to reduced updraft, whereas, during summer-monsoon season the aerosol effect is facilitated by favorable meteorological condition, due to strong updraft.

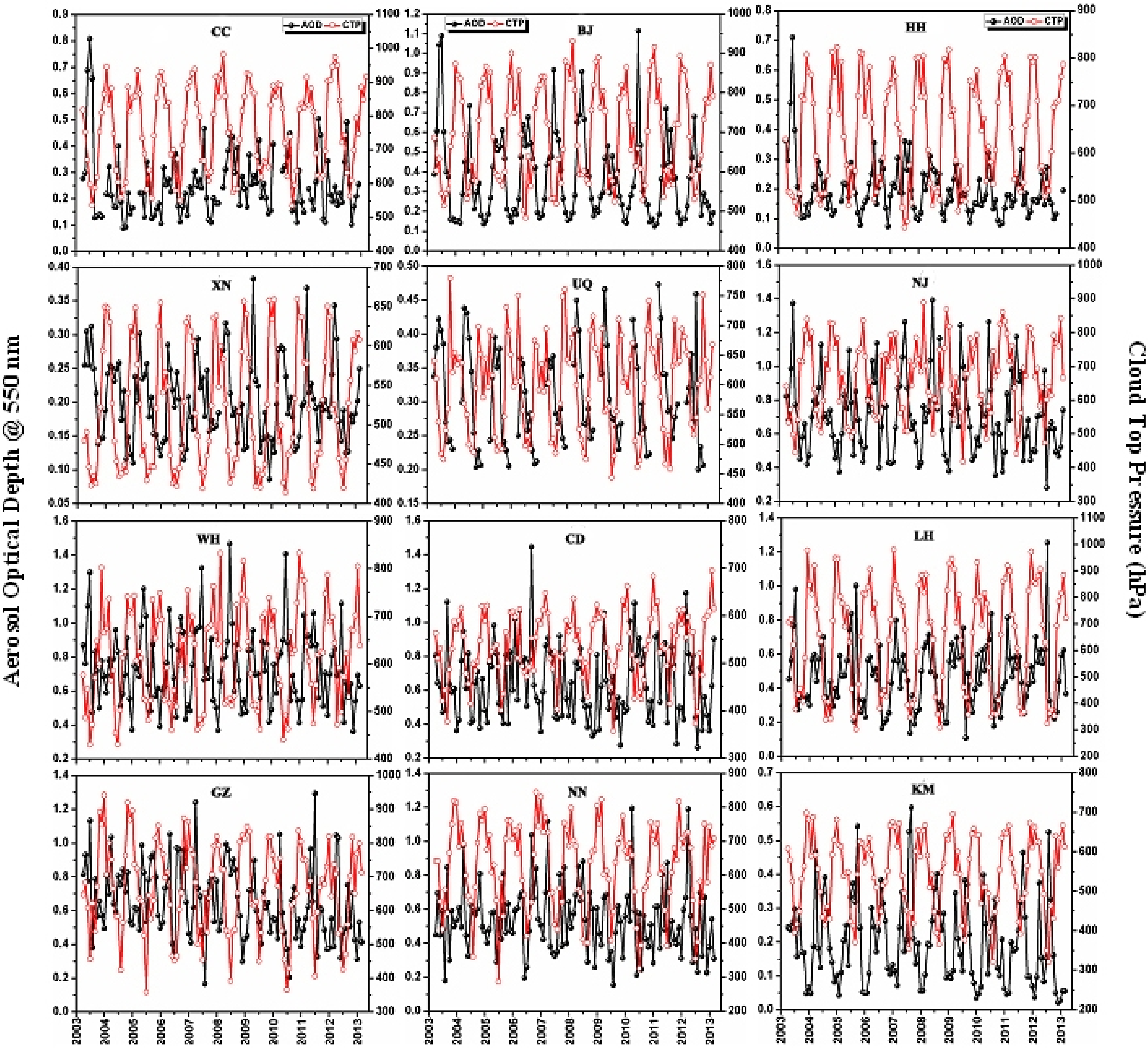

Fig. S2. Same as Fig. S1, but for cloud top pressure.

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