SUPPLEMENTARY MATERIAL

Aerosol-Cloud-Precipitation interactions over major cities in South Africa: Impact on regional environment and climate change

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SM-1. Validation reports of MODIS with ground-based data

The spatio-temporal data analysis carried out between satellite- and ground-based instruments help both in identifying the uncertainties of these retrievals and their local spatial behavior (Ichoku *et al.,* 2002). Previous studies have also used AERONET measured AODs to validate MODIS derived AODs. Chu *et al.* (2002) and Diner *et al.* (2001) performed validation over SA with a limited data for short period from July to September of 2000. Prasad *et al.* (2007) found a good correlation (R^2=0.47) between MODIS and AERONET over Kanpur during winter but a poor correlation (R^2=0.29) in summer. Gupta *et al.* (2013) compared MODIS derived AODs with AERONET derived AODs over Lahore and found a good correlation between the two (R^2=0.72). Recently, Bibi *et al.* (2015) reported over all good agreement between MODIS and AERONET AODs with correlation coefficients (R^2) of 0.71, 0.67, 0.76, and 0.61 over Karachi, Lahore, Jaipur, and Kanpur, respectively.

SM-2. Relationship between AOD and WV

In order to understand the aerosol impact and the process of the hydrological cycle, we investigated the change in column water vapour in relation to aerosols (Myhre *et al*., 2007). There are five near-infrared bands in MODIS just around water vapor band (940 nm) for remote sensing of clear sky column water vapour amount. It is so designed to observe water vapor absorption by solar radiation reflected by bottom surface at near-infrared. Through the use of the ratios of water absorbing bands with atmospheric window bands, the variation of surface reflectance with wavelength for most land surfaces is being removed (King *et al*., 1992). The spatial correlation of AOD_{550} with WV during clear sky is shown in Fig. SM-1a. It is found to be positive correlation (0.1-0.2) over Skukuza and Pretoria and negative correlation (>-0.3) over rest of the other locations.

The temporal correlation (Fig. SM-1b) between AOD_{550} and WV shows that both quantities only co-vary at the beginning of the year but they tend to have opposite trend in the rest of the year over all the locations. The correlation coefficients between for the above relationship observed to -0.463, 0.254, -0.464, 0.123, 0.461 and 0.054 for the locations Bloemfontein, Cape Town, Durban, Potchefstroom, Pretoria and Skukuza, respectively. It is evident that Pretoria has the highest correlation coefficient which agrees well with the time series plot indicates that summer months has higher correspondence between AOD_{550} and WV (Ranjan *et al.,* 2007). It may be also noted that aerosol influence the cloud formation over Pretoria as the location experiences higher rainfall during that season (Adesina *et al*., 2014). Aerosol may not be playing a significant role in cloud formation over Bloemfontein, Durban and Skukuza.

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). 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.

SM-3. Relationship between AOD and CTP

The spatial correlation between AOD and CTP presented in Fig. SM-2a shows that Durban, Potchefstroom and Bloemfontein have positive correlation, while the other locations observed to have negative correlation. CTP tends to have higher negative correlation at higher latitude as reported by some studies (Kaufman *et al*., 2005; Myhre *et al*., 2007; Sekiguchi *et al*., 2009) as in the case of Pretoria and Skukuza. These correlation facts may be caused by the large-scale meteorological variations. 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 (Kaufman *et al.,* 2005). 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. In the time series plot for the spatial averages (Fig. SM-2b), we can see same pattern of CTP increasing to a maximum value from January to July in all the stations, except Cape Town, where it first decreased to a minimum value in May then increased to July before it started to decrease at the end of the year. The correlation coefficients are observed to be Bloemfontein (0.260), Cape Town (0.182), Durban (0.203), Potchefstroom (-0.225), Pretoria (-0.452), and Skukuza (-0.327).

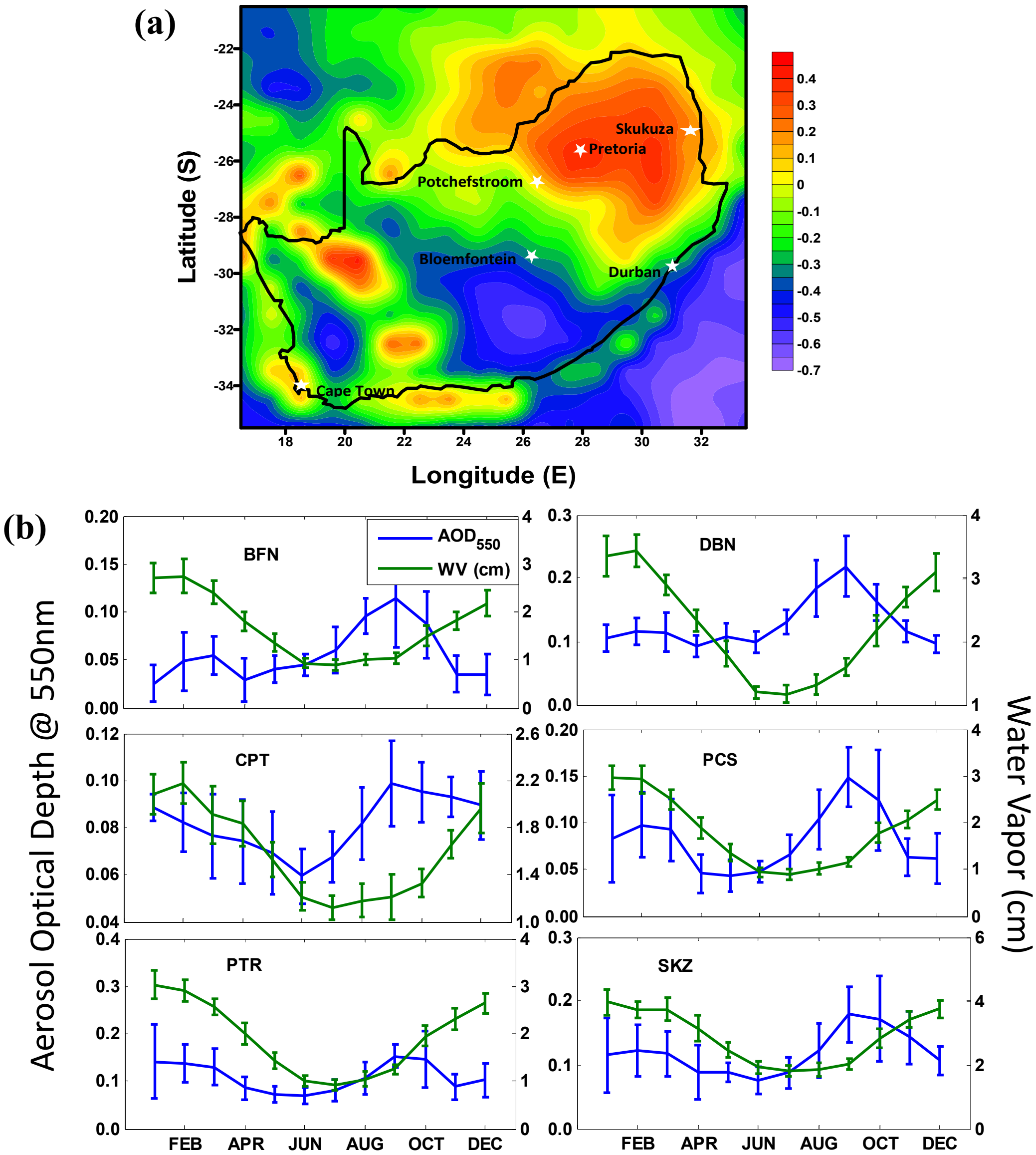

Fig. SM-1. (a) Spatial correlation map and (b) time series plots with standard deviation between AOD and WV over six locations in SA during 2004–2013.

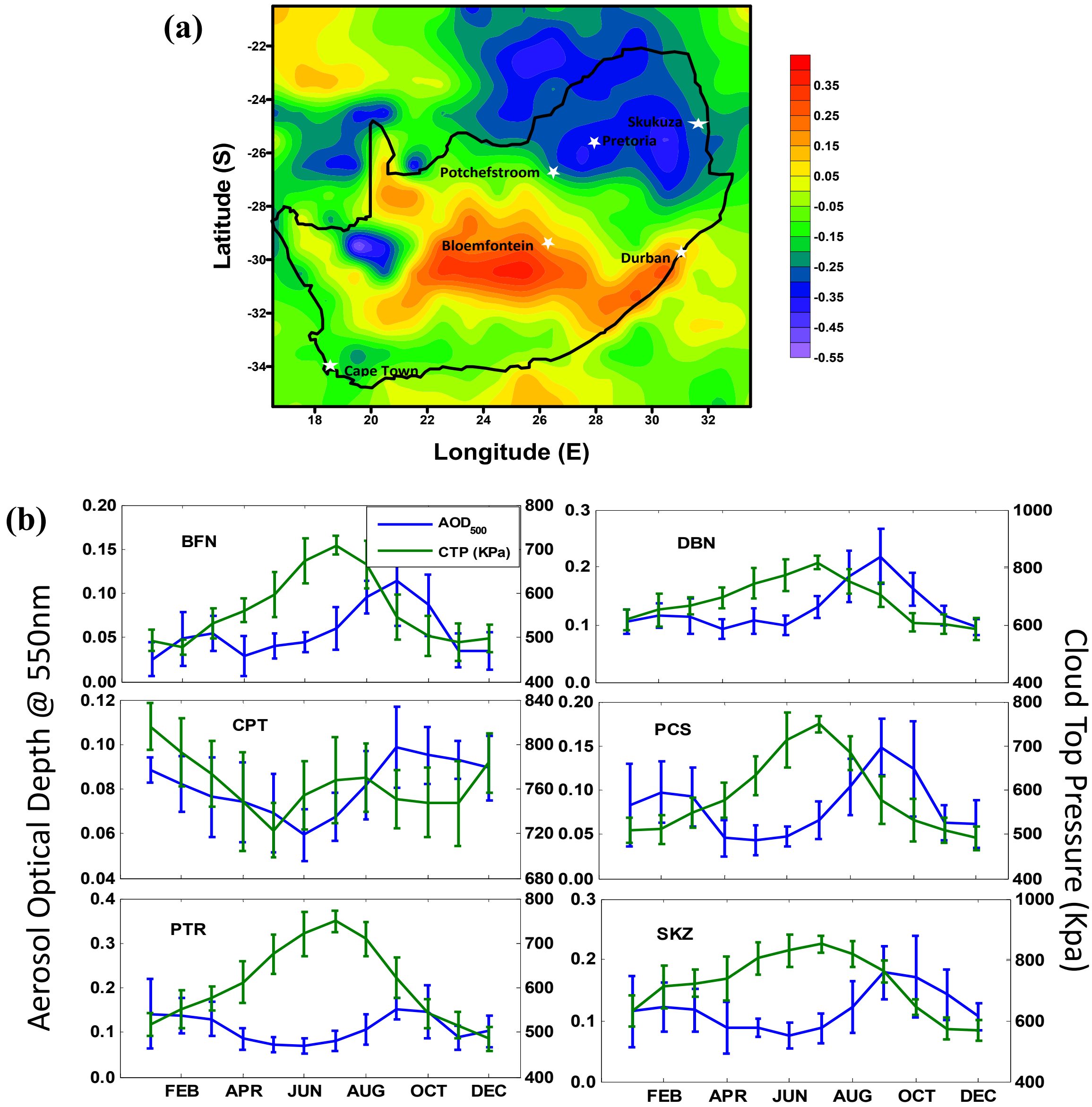

Fig. SM-2. Same as Fig. SM-1, but for AOD vs. CTP.

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