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A study of the impact of synoptic weather conditions and water vapor on aerosol-cloud relationships over major urban clusters of China

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Abstract. The relationships between aerosol optical depth (AOD), cloud cover (CC), and cloud top pressure (CTP) over three major urban clusters in China are studied under different sea level pressure (SLP) and water vapor (WV) regimes using a decade (2003-2013) of MODIS satellite-retrieved data. Over all urban clusters, for all SLP regimes, CC is found to increase with AOD, thus pointing out that the CC dependence on AOD cannot be explained by synoptic covariation, as approximated by SLP, alone. WV is found to have a stronger impact on CC than AOD. This impact is more pronounced at high aerosol load than at low aerosol load. Hence, studies of AOD-CC relationships, based on satellite data, will greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they will probably underestimate the AOD impact in regions where AOD and WV have opposite seasonal variations. Further, this impact shows that the hydrological cycle interferes with the aerosol climatic impact and we need to improve our understanding of this interference. Our results also suggest that studies attributing CTP long-term changes to changes in aerosol load might have a WV bias.

1 Introduction

Aerosols are known to impact the formation, optical properties, and life cycle of clouds (e.g., Ramanathan et al., 2001; Lohmann and Feichter, 2005; Tao et al., 2012; Boucher et al., 2013), either by increasing the cloud droplet number concentration and simultaneously decreasing the droplet size with a fixed water content, known as the first indirect effect (Twomey, 1974), or by suppressing precipitation formation, enhancing at the same time the cloud cover and cloud lifetime, known as the second indirect effect (Albrecht, 1989). In addition, by scattering or absorbing solar and terrestrial short-wave radiation (direct effect), aerosols affect temperature on the Earth's surface also perturbing the vertical temperature structure (Haywood and Boucher, 2000; Menon et al., 2002). Absorption in the atmosphere may also impact clouds by perturbing the vertical temperature structure (semi-direct effect, Ackerman et al., 2000). So, it is important to understand and quantify the microphysical impact of both natural and anthropogenic aerosols on clouds, in order to understand and predict climate change (Anderson et al., 2003; Forest et al., 2002; Knutti et al., 2002).

Urban clusters constitute a major political and economic issue in China. Increased numbers of cities of different sizes and intensive urbanization are prominent features in these regions, which extend over hundreds of kilometers. These city

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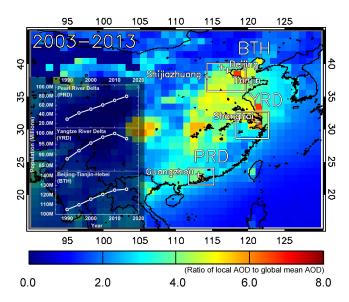


Figure 1. Map of China with the ratio of local AODs to the global mean AOD (\sim 0.1523) for the period 2003–2013. The position of the three urban clusters studied here (white squares, Beijing–Tianjin–Hebei: BTH, Yangtze River Delta: YRD, and Pearl River Delta: PRD) and their five major cities (white crosses) is marked. Estimates of the population (in millions, M) for the period 1990–2015 for the three urban clusters (CIESIN/CIAT/SEDAC, 2005) are also embedded in the map.

clusters are among the most dynamic and rapidly growing regions of China. Several such clusters have emerged in the past 2 decades and are still evolving. The city clusters studied here, namely the ones in the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Beijing-Tianjin-Hebei area (BTH), are the among the most rapidly growing, characterized by a spectacular population growth over the last 20 years (Fig. 1). These regions, with aerosol loads sometimes higher than the global average (Fig. 1), constitute extensive spatial sources of large quantities of aerosols as a result of human activities (industry, construction, traffic, etc.) and biomass burning, while occasionally transport of mineral dust from China's deserts adds to the aerosol burden of these regions (Zhao and Li, 2007; Jin and Shepherd, 2008). Moreover, the three regions exhibit significant climatic differences, driven also by the Asian monsoon, and hence they are suitable for the investigation of aerosol-cloud relations under different meteorological conditions.

The aim of this study is to study the influence of synoptic weather conditions and atmospheric water vapor amounts on aerosol optical depth(AOD)–cloud cover(CC) relationships, while at the same time obtain some insight on possible impacts on local climate that might result over the extended urbanization clusters of China due to aerosols. Towards this aim, we use 10 consecutive years (2003–2013) of AOD, CC, clear-sky water vapor (WV), and cloud top pressure (CTP) satellite data from MODIS TERRA and AQUA in conjunc-

tion with sea level pressure (SLP) from NCEP/NCAR Reanalysis data.

2 Data and methods

The three major urban clusters of China have been selected so as to be representative of three different climatic regions. China can be divided into five climate regions (Song et al., 2011), namely the temperate monsoon, the subtropical monsoon, the tropical monsoon, the temperate continental, and the plateau/mountain climate region. These are mainly influenced by the Asian monsoon systems and the Tibetan Plateau (Domros and Peng, 1988; Ye and Gao, 1979; Ding and Murakami, 1994). In particular, the Asian monsoon system has a major effect on the rainy seasons across the country. It starts with the pre-monsoonal rain period over south China in early April and lasts from May until August. The summer monsoon rain belt propagates northward to the Yangtze River basin in June and finally to northern China in July. In August, when the monsoon period ends, the rain belt moves back to southern China. Due to the migration of the monsoon across China, the length of the rain season differs between southern and northern China (Song et al., 2011). In particular, the BTH urban cluster is a temperate monsoon climate region, while the YRD urban cluster is a subtropical monsoon climate region, and the PRD urban cluster is a tropical monsoon climate region (Fig. 1). The BTH domain (35.5– 40.5° N, 113.5–120.5° E) is an area with rapid industrial and economic development, reflected also at the high AOD levels (more than 4 times the global average) over the region (Fig. 1). The YRD domain (28.5–33.5° N, 117.5–123.5° E), is an area with significant black carbon (Streets et al., 2001; Bond et al., 2004) and sulfate (Lu et al., 2010) emissions. Finally, the PRD domain (21.5–24.5° N, 111.5–115.5° E) is an area within the intertropical convergence zone (ITCZ) migration belt, with high anthropogenic aerosol emissions (Streets et al., 2003, 2008; Lei et al., 2011). Over the three regions of interest and within the study period, only weak overall upward trends have been reported (Guo et al., 2011).

Aerosol and cloud parameters from the MODIS instrument aboard the TERRA and AQUA satellites (collection 5.1, level 3, $1^{\circ} \times 1^{\circ}$ daily data) for the period 2003–2013 are used in this study. In particular, to investigate aerosol–cloud interactions, we use aerosol optical depth at 550 nm (AOD₅₅₀ or just AOD), CC, WV for clear conditions (Remer et al., 2005, 2008; King et al., 2003) and CTP from both satellites. Aerosol index (AI), defined as the product of the AOD and Ångström exponent, is a good proxy to quantify cloud condensation nuclei and has been applied in many previous aerosol–cloud interaction (ACI) studies (e.g., Costantino and Bréon, 2010 (off-coast Namibia and Angola)). However, in the present study the use of AI would not be appropriate, because our study is conducted over land areas. This has to do with the use of the Ångström exponent in the derivation of

AI, namely, the Ångström exponent is not reliable over land areas. We quote a personal communication with L. Remer (20 June 2010), NASA GSFC: "Ångström over land is not reliable and we recommend strongly not to use it"; hence, AOD is used in our study. Additionally, to examine the aerosol–cloud interactions under different meteorological conditions, such as low and high pressure systems, we used daily sea level pressure (SLP) data from the NCEP/NCAR Reanalysis for the same period. The original $2.5^{\circ} \times 2.5^{\circ}$ NCEP/NCAR SLP data were regridded using bilinear interpolation in order to match the MODIS $1^{\circ} \times 1^{\circ}$ level 3 data set.

Considering that meteorological conditions may have an impact on satellite derived aerosol-cloud relationships, and to investigate the influence of synoptic meteorological conditions on the AOD-CC relationship, the AOD, CC, WV and CTP MODIS data were classified into three SLP classes (less than 1008 hPa for low pressure systems, between 1008 and 1017 hPa, and finally greater than 1017 hPa for high pressure systems) using NCAR/NCEP SLP data, and also according to atmospheric WV quantities. This was done as follows, for each of the three urban clusters studied: concurrent MODIS AOD, WV, CC and CTP values were assigned to one of the three SLP classes according to the concurrent NCAR/NCEP sea level pressure. Then, within each of the three SLP subsets, containing each time series of concurrent AOD, WV, CC, and CTP values, the data were binned in equally sized bins (thus non-equal sample size bins, as this would make the comparison between the three clusters difficult) according to AOD and WV. This resulted in 100 bins (10 AOD bins for AOD between 0 and 1, bin step $0.1 \times 10 \text{ WV}$ bins for WV between 0 and 10 cm, bin step 1). The mean of the CC, and CTP values corresponding to AOD and WV within each bin was then calculated (in case there were more than six values of the respective variable within the studied bin). The same was repeated once more using AOD and CC equally sized bins. This resulted in 100 bins (10 AOD bins for AOD between 0 and 1, bin step 0.1×10 CC bins for CC between 0 and 1, bin step 0.1). The mean of the WV values corresponding to AOD and CC within each bin was then calculated (in case there were more than six values of WV within the studied bin).

3 Results and discussion

To gain an insight into the levels, trends, interannual variability, and seasonal variation of AOD and CC over the study regions, we first examined the time series of AOD, CC, CTP, and WV from MODIS TERRA and AQUA satellites over five grid points where cities of the three major urban clusters under study are located, for the period 2003–2013 (Figs. S1 and S2 in the Supplement). The results from both satellites are similar, with the highest values of AOD, CC, and WV occurring during the summer months, while CTP is higher during winter and lower during summer over all five cities

(i.e., cloud top height, hereafter denoted as CTH, also peaks in summer). The majority of the AOD values over the BTH urban cluster are between 0.3 and 1.4, while over YRD they are between 0.5 and 1.3, and between 0.5 and 1 over the PRD urban cluster. BTH, with an average AOD $_{550}$ of 0.654 ± 0.15 during the study period (2003–2013), experiences somewhat heavier aerosol loading than the other two regions with average AODs of 0.646 ± 0.18 (YRD) and 0.590 ± 0.16 (PRD). Further, as we move from north to south, CC and WV increases, while CTP variability also increases. Additionally, in the variables we will use in this study, no major trends are apparent during the study period (Figs. S1 and S2).

To investigate the influence of synoptic meteorological conditions on the AOD–CC relationship (Fig. S3), and to exclude, at least partially, artifacts on the AOD–CC relationship resulting from synoptically induced covariance (Mauger and Norris, 2007; Loeb and Schuster, 2008; Quaas et al., 2010; Gryspeerdt et al., 2014a), the MODIS data were classified into three SLP classes (from NCAR/NCEP data, see above) and the AOD–CC–WV relationship was examined at the low and high SLP classes.

Figures 2–4 show the AOD–CC–WV relationship over the three urban clusters studied for two of the three SLP classes, namely low SLP (SLP < 1008 hPa, Figs. 2-4a and b) and high SLP (SLP>1017 hPa, Figs. 2-4e and f), as the meteorological conditions for these two classes are more clearly defined. The SLP < 1008 hPa class is representative of the core of low pressure systems and hence of atmospheric circulation typical of these systems (e.g., ascending motions of air). The SLP>1017 hPa class is representative of the core of high pressure systems and hence of atmospheric circulation typical of these systems (e.g., descending motions of air). Furthermore, the low and high SLP systems are completely different in terms of horizontal transport patterns. The 1008 hPa < SLP < 1017 hPa class is less clearly defined in terms of atmospheric conditions, since it might contain meteorological conditions typical of the periphery of low pressure systems or typical of the periphery of high pressure systems (e.g., troughs, ridges), and hence it is omitted from the discussion (figure for 1008 < SLP < 1017 is also available, but not shown here). Figures on the left present results in bins, while figures on the right present results as line graphs. Water vapor is in 1 cm bins and AOD is in 0.1 bins. The same analysis was also performed with MODIS TERRA data (Figs. S4 to S6), and the results are qualitatively and to a large part also quantitatively in accord with the MODIS AQUA ones. With increasing SLP, the amount of WV in the atmosphere decreases (Figs. 2a, b, e, and f; 3a, b, e, and f; and 4a, b, e, and f). This is due to the fact that the majority of available AOD-CC retrieval pairs for the low SLP class occurs during summer, when also the majority of available AOD-CC retrieval pairs for WV > 3 cm occurs (Figs. S7 and S8). Wang et al. (2015) also noted the different humidity levels during summer and winter over east China. Additionally, as low SLP synoptic systems are associated with updrafts, the occurrence

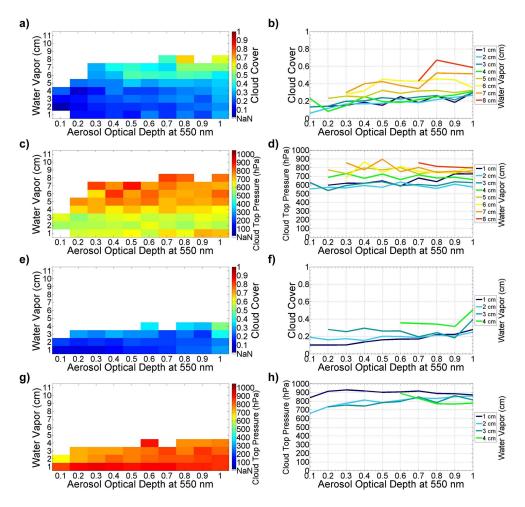


Figure 2. MODIS AQUA data for the Beijing–Tianjin–Hebei (BTH) urban cluster, 2003–2013, AOD-WV-CC (**a–b**), AOD-WV-CTP (**c–d**) for SLP < 1008 hPa, and AOD-WV-CC (**e–f**), AOD-WV-CTP (**g–h**) for SLP > 1017 hPa. NaN on the cloud data color bar denotes no values or fewer than six values in this bin. Figures on the left present average CC and CTP values in 1 cm WV and 0.1 AOD bins, while figures on the right present results as line graphs. The CC–AOD line graphs and CTP–AOD relations were calculated by averaging CC and CTP within 0.1 AOD bins for several 1 cm WV classes.

of these systems in summer, when land and sea temperatures, and hence also evaporation, are higher, more WV can be transported up in the atmosphere. Other authors have also noted the correlation of AOD with WV. For example, Alam et al. (2010), report positive AOD-WV correlation over Pakistan due to their common seasonal patterns. On the other hand, Balakrishnaiah et al. (2012) report positive AOD-WV correlation over India but negative correlation over some Indian Ocean regions. It is evident that WV has a strong impact on CC, perhaps even stronger than the AOD impact on CC (Figs. 2–4a, b, e, and f). In fact, over PRD the impact of AOD on CC for constant WV seems negligible (Fig. 4a, b, e, and f). In the other two regions, BTH and YRD, CC might increase by up to 0.1 at the most as AOD increases from 0.2 to 1 under constant WV, while CC might increase by up to 0.4 for WV increases from 1 to 8 cm under constant AOD. For detailed statistics of the AOD-CC and AOD-CTP relationships please refer to Tables S1 to S5 of the Supplement. Also, for the response of CC to AOD in terms of seasonality, given the strong seasonal variability in aerosol and cloud shown in Figs. S1 and S2, please refer to Tables S1 and S2 and Fig. S3.

Hence, studies of AOD–CC relationships based on satellite data, that do not take WV into account, will greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations. Keeping in mind the reasons for the observed overestimations in the three regions studied here, it is logical to infer that, in regions where AOD and WV have opposite seasonal variations, the AOD impact on CC may most likely be underestimated if WV is not taken into account. This result is in agreement with recent results from other authors that noted the large possible impact of different meteorological variables on AOD–CC relationships (e.g., Mauger and Norris, 2007; Quaas et al., 2010; Koren

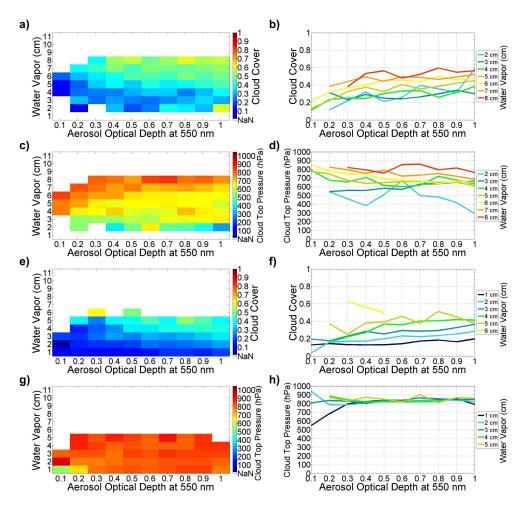


Figure 3. As in Fig. 2, but for the Yangtze River Delta (YRD) urban cluster.

et al., 2010; Engström and Ekman, 2010; Chand et al., 2012; Grandey et al., 2013). Most importantly, it is in agreement with recent reports that have given qualitative indications that water vapor (Ten Hoeve et al., 2011) or relative humidity (Loeb and Schuster, 2008; Koren et al., 2010; Grandey et al., 2013) might have a strong influence on AOD-CC relationships. We also note that despite the remarks made above, even after accounting for WV and synoptic variability as manifested by SLP, weakened positive relationships between AOD and CC often remain (Figs. 2-4a, b, e and f), although this impact in our study regions is much smaller than the one that would have been estimated ignoring synoptic and WV variability. In fact, in the three areas of study, where AOD and WV have similar seasonal variations, if the water vapor effect is taken into account, the slopes of the CC-AOD relationship for AOD > 0.2 might be reduced by up to 90%. We suggest that these results should be taken into consideration in future studies trying to explain the weekly cycles of cloud cover and other meteorological parameters (e.g., temperature, solar radiation, precipitation) observed in some regions of the planet, through the weekly working cycle and the indirect effects of aerosols (e.g., Georgoulias et al., 2015). Also, the results suggest a profound interference of the hydrological cycle with the aerosol climatic impact; this needs further investigation. Recent studies also point out different aspects of the aforementioned interference (Grandey et al., 2014; Rosenfeld et al., 2014; Gryspeerdt et al., 2015,).

In all SLP and urban cluster cases, there is no apparent systematic increase of AOD with WV, and it does not appear that increased WV is systematically associated with large increases in AOD (Figs. 5 and 6). This indicates that there is no large systematic AOD increase at increased WV. Further, it is apparent that the largest part of the differences in the AOD–CC slope between low and high SLP synoptic conditions is due to the differences in WV between these conditions (see Fig. 5, Figs. S7, and S8, and also compare parts b and f of Figs. 2–4).

CTP is a cloud parameter that can be used as a proxy for cloud vertical development; hence a number of recent studies have investigated its role in AOD–CC interactions over the region of eastern Asia (e.g., Kumar, 2013; Alam et al., 2014; Tang et al., 2014; Wang et al., 2014) and glob-

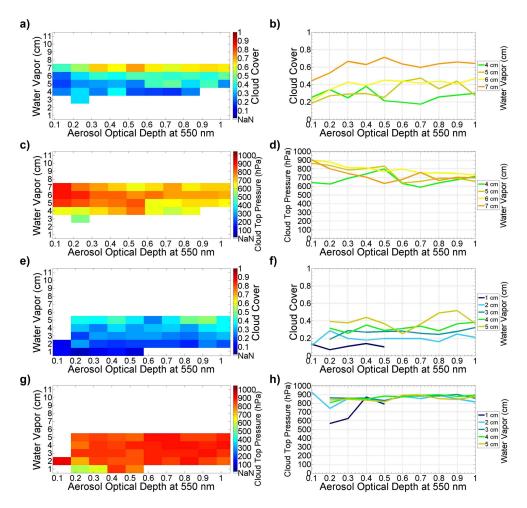


Figure 4. As in Fig. 2, but for the Pearl River Delta (PRD) urban cluster.

ally (e.g., Gryspeerdt et al., 2014a). Recently, Gryspeerdt et al. (2014b), using satellite data, reported that apart from AOD, CTP is also strongly correlated to CTP and argue that influences such as aerosol humidification and meteorology play an important role and should be considered in studies of aerosol-cloud interactions. CTP variations over the study areas were not dominantly driven by AOD, irrespective of the pressure system and WV bins (Figs. 2-4c, d, e, and h). However, at low SLP regimes, CTP decreased with AOD over PRD, much less so over YRD, and was not impacted by AOD over BTH (Figs. 2–4c and d), while at high SLP it was not impacted by AOD in all three urban clusters (Figs. 2-4e and h). Finally, CTP was found to increase considerably with WV content only at low SLP over BTH and YRD. Hence, studies attributing CTP, CTH, or cloud top temperature (CTT) long-term changes to changes in aerosol load, without accounting for this WV effect (e.g., Devasthale et al., 2005), might lead to wrong quantifications. Although differences between MODIS AQUA and TERRA (Meskhidze et al., 2009) are outside the scope of this study, we mention briefly that CTP from AQUA is lower than TERRA over all

regions and under all pressure systems (compare Figs. 2 to 4 with Figs. S4 to S6), which is possibly due to the fact that clouds are more well-developed in the afternoon (AQUA overpass) than in the morning (TERRA overpass).

4 Conclusions

In this work, we used a decade (2003–2013) of aerosol and cloud parameters from the MODIS instrument to investigate the aerosol–cloud interactions over three major urban clusters of China, representative of three different climatic regions. We investigated the AOD–CC relationship under different synoptic conditions using SLP data, and under different clear-sky WV contents. Over all urban clusters, and for all SLP regimes, CC is found to increase with AOD, thus pointing out that the CC dependence on AOD cannot be explained by synoptic covariation, as approximated by SLP, alone. It is found that at AOD > 0.2, the AOD impact on CC at low SLP conditions is about 2 times higher than its impact at high SLP conditions. Further, at its largest part, this difference is due to

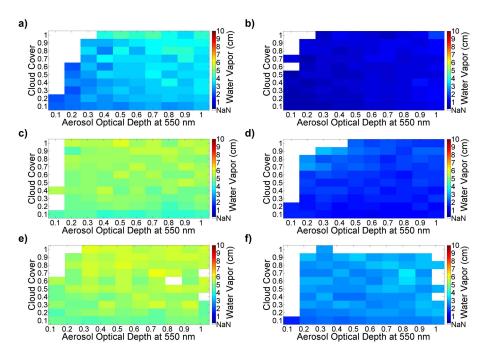


Figure 5. MODIS AQUA mean WV amounts for 0.1 AOD and 0.1 CC bins over the (BTH) (top), YRD (middle), and PRD (bottom) urban clusters for 2003–2013, for SLP < 1008 hPa (left) and SLP > 1017 hPa (right). NaN on the cloud data color bar denotes no values or fewer than six values in this bin.

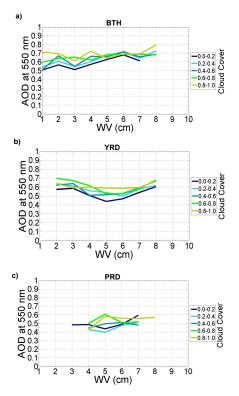


Figure 6. MODIS AQUA, AOD-WV-CC for SLP < 1008 hPa over (a) Beijing—Tianjin—Hebei (BTH) urban cluster, (b) Yangtze River Delta urban cluster (YRD), and (c) Pearl River Delta (PRD), 2003—2013. The AOD—WV line graphs were calculated by averaging AOD and WV within 1cm WV bins for several 0.2 CC classes.

WV differences between low and high SLP conditions, rather than arising from differences in horizontal transport patterns; hereupon we stratified the data into three SLP bins to examine AOD–CC–WV relationships under different pressure systems. In most cases, WV is found to be constant with increasing AOD loading, while there is a positive relationship between cloud cover and water vapor for fixed AOD. Moreover, the AOD–CC relationship is positive under all pressure conditions.

In general, WV has a strong impact on CC and thus, studies of aerosol-cloud interactions based on satellite data that do not account for this parameter, may result in erroneous quantitative and qualitative results; namely, studies of AOD-CC relationships, based on satellite data, will greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they may probably greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations. In the three areas of study, where AOD and WV have similar seasonal variations, if the water vapor effect is taken into account, the slopes of the CC-AOD relationship for AOD > 0.2 might be reduced by up to 90%. Further, this WV impact on AOD-CC relationships shows that the hydrological cycle interferes with the aerosol climatic impact and we need to improve our understanding of this interference.

In addition, cloud top pressure (CTP) at low SLP regimes is found to decrease more with AOD over the PRD and much less so over the YRD urban cluster, while there is no significant impact by AOD over BTH. On the other hand, at

high SLP regimes, AOD does not seem to impact CTP significantly. Finally, over the BTH and YRD urban clusters, CTP is found to increase considerably with increasing WV, only at low SLP synoptic regimes. Similar to the case of AOD–CC relations, these results suggest that studies trying to relate CTP, CTH, and CTT changes with changes in aerosol load should account for this WV effect.

It is also found that there is no large systematic AOD increase at high WV.

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References

- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of Tropical Cloudiness by Soot, Science, 288, 1042–1047, doi:10.1126/science.288.5468.1042, 2000.
- Alam, K., Iqbal, M. J., Blaschke, T., Qureshi, S., and Khan, G.: Monitoring spatio-temporal variations in aerosols and aerosolcloud interactions over Pakistan using MODIS data, Adv. Space Res., 46, 1162–1176, 2010.
- Alam, K., Khan, R., Blaschke, T., and Mukhtiar, A.: Variability of aerosol optical depth and their impact on cloud properties in Pakistan, J. Atmos. Sol.-Terr. Phy., 107, 104–112, doi:10.1016/j.jastp.2013.11.012, 2014.
- Albrecht, B. A: Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227–1230, doi:10.1126/science.245.4923.1227, 1989.
- Anderson, T. L., Charlson, R. J., Schwartz, S. E., Knutti, R., Boucher, O., Rodhe, H., and Heintzenberg, J.: Climate forcing by aerosols-a hazy picture, Science, 300, 1103, doi:10.1126/science.1084777, 2003.

- Balakrishnaiah, G., Raghavendra Kumar, K., Suresh Kumar Reddy, B., Rama Gopal, K., Reddy, R. R., Reddy, L. S. S., Swamulu, C., Nazeer Ahammed, Y., Narasimhulu, K., KrishnaMoorthy, K., and Suresh Babu, S.: Spatio-temporal variations in aerosol optical and cloud parameters over Southern India retrieved from MODIS satellite data, Atmos. Environ., 47, 435– 445, 2012.
- Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J. H., and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi:10.1029/2003JD003697, 2004.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and aerosols, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge Univ. Press, Cambridge, UK, New York, NY, USA, 573–632, 2013.
- Chand, D., Wood, R., Ghan, S. J., Wang, M., Ovchinnikov, M., Rasch, P. J., Miller, S., Schichtel, B., and Moore, T.: Aerosol optical depth increase in partly cloudy conditions, J. Geophys. Res., 117, D17207, doi:10.1029/2012JD017894, 2012.
- CIESIN/CIAT/SEDAC: Gridded Population of the World Version 3 (GPWv3): Population Grids, Palisades, Socioeconomic Data and Applications Center (SEDAC), Columbia University, NY, available at: http://sedac.ciesin.columbia.edu/gpw (last access: 11 December 2014), 2005.
- Costantino, L. and Bréon, F.-M.: Analysis of aerosol-cloud interaction from multi-sensor satellite observations, Geophys. Res. Lett., 37, L11801, doi:10.1029/2009GL041828, 2010.
- Devasthale, A., Krüger, O., and Graßl, H.: Change in cloud top temperatures over Europe, IEEE Geosci. Remote S., 2, 333–336, 2005
- Ding, Y. H. and Murakami, M.: The Asian Monsoon, China Meteorological Press, Beijing, China, 1994.
- Domros, M. and Peng, G.: The Climate of China, Springer Verlag, Berlin, 1988.
- Engström, A. and Ekman, A. M.: Impact of meteorological factors on the correlation between aerosol optical depth and cloud fraction, Geophys. Res. Lett., 37, L18814, doi:10.1029/2010GL044361, 2010.
- Forest, C. E., Stone, P. H., Sokolov, A. P., Allen, M. R., and Webster, M. D.: Quantifying uncertainties in climate system properties with the use of recent climate observations, Science, 295, 113–117, doi:10.1126/science.1064419, 2002.
- Georgoulias, A. K., Kourtidis, K. A., Alexandri, G., Rapsomanikis, S., and Sanchez-Lorenzo, A.: Common summertime total cloud cover and aerosol optical depth weekly variabilities over Europe: sign of the aerosol indirect effects?, Atmos. Res., 153, 59–73, doi:10.1016/j.atmosres.2014.07.031, 2015.
- Grandey, B. S., Stier, P., and Wagner, T. M.: Investigating relationships between aerosol optical depth and cloud fraction using satellite, aerosol reanalysis and general circulation model data, Atmos. Chem. Phys., 13, 3177–3184, doi:10.5194/acp-13-3177-2013, 2013.

- Grandey, B. S., Gururaj, A., Stier, P., and Wagner, T. M., Rainfall–aerosol relationships explained by wet scavenging and humidity, Geophys. Res. Lett., 41, 5678–5684, doi:10.1002/2014GL060958, 2014.
- Gryspeerdt, E., Stier, P., and Partridge, D. G.: Satellite observations of cloud regime development: the role of aerosol processes, Atmos. Chem. Phys., 14, 1141–1158, doi:10.5194/acp-14-1141-2014, 2014a.
- Gryspeerdt, E., Stier, P., and Grandey, B. S.: Cloud fraction mediates the aerosol optical depth-cloud top height relationship, Geophys. Res. Lett., 41, 3622–3627, doi:10.1002/2014GL059524, 2014b.
- Gryspeerdt, E., Stier, P., White, B. A., and Kipling, Z.: Wet scavenging limits the detection of aerosol effects on precipitation, Atmos. Chem. Phys., 15, 7557–7570, doi:10.5194/acp-15-7557-2015, 2015.
- Guo, J. P., Zhang, X. Y., Wu, Y. R., Zhaxi, Y., Che, H. Z., La, B., Wang, W., and Li, X. W.: Spatio-temporal variation trends of satellite-based aerosol optical depth in China during 1980–2008, Atmos. Environ., 45, 6802–6811, doi:10.1016/j.atmosenv.2011.03.068, 2011.
- Haywood, J. M. and Boucher, O.: Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review, Rev. Geophys., 38, 513–543, doi:10.1029/1999RG000078, 2000.
- Jin, M. and Shepherd, J. M.: Aerosol relationships to warm season clouds and rainfall at monthly scales over east China: urban land versus ocean, J. Geophys. Res., 113, D24590, doi:10.1029/2008JD010276, 2008.
- King, M. D, Menzel, W. P., Kaufman, Y. J., Tanré, D., Gao, B.-C., Platnick, S., Ackerman, S. A., Remer, L. A., Pincus, R., and Hubanks, P. A.: Cloud and aerosol properties, precipitable water, and profiles of temperature and water vapor from MODIS, IEEE Geosci. Remote S., 41, 442–458, doi:10.1109/TGRS.2002.808226, 2003.
- Knutti, R., Stocker, T. F., Joos, F., and Plattner, G. K.: Constraints on radiative forcing and future climate change from observations and climate model ensembles, Nature, 416, 719–723, doi:10.1038/416719a, 2002.
- Koren, I., Feingold, G., and Remer, L. A.: The invigoration of deep convective clouds over the Atlantic: aerosol effect, meteorology or retrieval artifact?, Atmos. Chem. Phys., 10, 8855–8872, doi:10.5194/acp-10-8855-2010, 2010.
- Kumar, A.: Variability of aerosol optical depth and cloud parameters over North Eastern regions of India retrieved from MODIS satellite data, J. Atmos. Sol.-Terr. Phy., 100, 34–49, doi:10.1016/j.jastp.2013.03.025, 2013.
- Lei, Y., Zhang, Q., He, K. B., and Streets, D. G.: Primary anthropogenic aerosol emission trends for China, 1990–2005, Atmos. Chem. Phys., 11, 931–954, doi:10.5194/acp-11-931-2011, 2011.
- Loeb, N. G. and Schuster, G. L.: An observational study of the relationship between cloud, aerosol and meteorology in broken low-level cloud conditions, J. Geophys. Res., 113, D14214, doi:10.1029/2007JD009763, 2008.
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715–737, doi:10.5194/acp-5-715-2005, 2005.
- Lu, Z., Streets, D. G., Zhang, Q., Wang, S., Carmichael, G. R., Cheng, Y. F., Wei, C., Chin, M., Diehl, T., and Tan, Q.: Sulfur dioxide emissions in China and sulfur trends in East Asia since

- 2000, Atmos. Chem. Phys., 10, 6311–6331, doi:10.5194/acp-10-6311-2010, 2010.
- Mauger, G. S. and Norris, J. R.: Meteorological bias in satellite estimates of aerosol-cloud relationships, Geophys. Res. Lett., 34, L16824, doi:10.1029/2007GL029952, 2007.
- Menon, S., Hansen, J., Nazarenko, L., and Luo, Y.: Climate effects of black carbon aerosols in China and India, Science, 297, 2250– 2253, doi:10.1126/science.1075159, 2002.
- Meskhidze, N., Remer, L. A., Platnick, S., Negrón Juárez, R., Lichtenberger, A. M., and Aiyyer, A. R.: Exploring the differences in cloud properties observed by the Terra and Aqua MODIS Sensors, Atmos. Chem. Phys., 9, 3461–3475, doi:10.5194/acp-9-3461-2009, 2009.
- Quaas, J., Stevens, B., Stier, P., and Lohmann, U.: Interpreting the cloud cover aerosol optical depth relationship found in satellite data using a general circulation model, Atmos. Chem. Phys., 10, 6129–6135, doi:10.5194/acp-10-6129-2010, 2010.
- Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, Science, 294, 2119–2124, doi:10.1126/science.1064034, 2001.
- Remer, L. A., Kaufman, Y. J., Tanré, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R.-R., Ichoku, C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B. N.: The MODIS Aerosol algorithm, products, and validation, J. Atmos. Sci., 62, 947–973, doi:10.1175/JAS3385.1, 2005.
- Remer, L. A., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanré, D., Mattoo, S., Martins, J. V., Ichoku, C., Koren, I., Yu, H., and Holben, B. N.: Global aerosol climatology from the MODIS satellite sensors, J. Geophys. Res., 113, D14S07, doi:10.1029/2007JD009661, 2008.
- Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., de Leeuw, G., Donovan, D. P., Kahn, R., Kinne, S., Kivekäs, N., Kulmala, M., Lau, W., Schmidt, K. S., Suni, T., Wagner, T., Wild, M., and Quaas, J.: Global observations of aerosol-cloud-precipitation-climate interactions, Rev. Geophys., 52, 750–808, doi:10.1002/2013RG000441, 2014.
- Song, Y., Achberger, C., and Linderholm, H. W.: Rain-season trends in precipitation and their effect in different climate regions of China during 1961–2008, Environ. Res. Lett., 6, 034025, doi:10.1088/1748-9326/6/3/034025, 2011.
- Streets, D. G., Gupta, S., Waldhoff, S. T., Wang, M. Q., Bond, T. C., and Yiyun, B.: Black carbon emissions in China, Atmos. Environ., 35, 4281–4296, doi:10.1016/S1352-2310(01)00179-0, 2001.
- Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108, 8809, doi:10.1029/2002JD003093, 2003.
- Streets, D. G., Yu, C., Wu, Y., Chin, M., Zhao, Z., Hayasaka, T., and Shi, G.: Aerosol trends over China, 1980–2000, Atmos. Res., 88, 174–182, doi:10.1016/j.atmosres.2007.10.016, 2008.
- Tang, J., Wang, P., Mickley, L. J., Xia, X., Liao, H., Yue, X., Sun, L., and Xia, J.: Positive relationship between liquid cloud droplet effective radius and aerosol optical depth over Eastern China from satellite data, Atmos. Environ., 84, 244–253, doi:10.1016/j.atmosenv.2013.08.024, 2014.

- Tao, W.-K., Chen, J.-P., Li, Z., Wang, C., and Zhang, C.: Impact of aerosols on convective clouds and precipitation, Rev. Geophys., 50, RG2001, doi:10.1029/2011RG000369, 2012.
- Ten Hoeve, J. E., Remer, L. A., and Jacobson, M. Z.: Microphysical and radiative effects of aerosols on warm clouds during the Amazon biomass burning season as observed by MODIS: impacts of water vapor and land cover, Atmos. Chem. Phys., 11, 3021–3036, doi:10.5194/acp-11-3021-2011, 2011.
- Twomey, S.: Pollution and the planetary albedo, Atmos. Environ., 8, 1251–1256, doi:10.1016/j.atmosenv.2007.10.062, 1974.
- Ye, D. Z. and Gao, Y. X.: The Meteorology of the Qinghai-Xizang Plateau, Chinese Science Press, Beijing, China, 1979.
- Wang, F., Guo, J., Wu, Y., Zhang, X., Deng, M., Li, X., Zhang, J., and Zhao, J.: Satellite observed aerosol-induced variability in warm cloud properties under different meteorological conditions over eastern China, Atmos. Environ., 84, 122–132, doi:10.1016/j.atmosenv.2013.11.018, 2014.

- Wang, F., Guo, J., Zhang, J., Huang, J., Min, M., Chen, T., Liu, H., Deng, M., and Li, X.: Multi-sensor quantification of aerosolinduced variability in warm clouds over eastern China, Atmos. Environ., 113, 1–9, doi:10.1016/j.atmosenv.2015.04.063, 2015.
- Zhao, F. and Li, Z.: Estimation of aerosol single scattering albedo from solar direct spectral radiance and total broadband irradiances measured in China, J. Geophys. Res., 112, D22503, doi:10.1029/2006JD007384, 2007.