Response to Referee #1

General comments

We thank the referee for his positive comments.

Major comment: Interpretation and communication of results

The referee agrees with our statement that "studies of AOD-CC relationships based on satellite data that do not take into account WV, might greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations" but is not convinced that the results show that "studies of AOD-CC relationships based on satellite data that do not take into account WV […] might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations". The reason is that none of the studied regions exhibit the proposed underestimation effect. However, we believe that although our study was limited to regions of similar AOD and WV seasonal variations, it nevertheless shows (as the referee agrees) that in regions of similar AOD and WV seasonal variations AOD-CC relationships become much weaker when WV is taken into account. The above is a consequence, as the figures 2-4 show, of the water vapor impact on CC and the fact that if WV is not accounted for, its impact appears as AOD impact due to the common AOD-WV seasonal cycle in the three regions of the present study. We believe that as a logical consequence it follows that, CC in regions where AOD and WV have opposite seasonal variations, AOD-CC relationships based on satellite data that do not take into account WV, will underestimate the AOD impact on CC (or, if you prefer, "will most probably underestimate the AOD impact on CC". Hence, we opt not to delete the respective part of the sentence, but rephrase it slightly as follows:

p14008.9-10, from "Hence, studies of AOD-CC relationships based on satellite data, might greatly overestimate or underestimate the AOD impact on CC in regions where AOD and WV have similar or opposite seasonal variations, respectively" to "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".

p14013.21, from "Hence, studies of AOD-CC relationships based on satellite data that do not take into account WV, might greatly overestimate the AOD impact on CC in regions where

AOD and WV have similar seasonal variations, while they might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations" to "Hence, studies of AOD-CC relationships based on satellite data that do not take into account WV, 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".

p14015.27, from "Namely, studies of AOD-CC relationships based on satellite data, might greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations" to "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 propably greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations".

The referee considers it misleading to imply in p14014.3 that "AOD does have an impact on CC even if synoptic and WV variability are accounted for" because "*not all synoptic variability has been accounted for and the remaining positive AOD-CC relationships do not imply a causal relationship*". Along these lines, the referee suggests rephrasing to "*even after accounting for SLP and WV, weakened positive relationships between AOD and CC often remain*". Along the referee line of thought, but taking also into account our argumentation about using SLP as a synoptic conditions proxy in p14012.7-21, we rephrase to "even after accounting for WV and synoptic variability as manifested by SLP, weakened positive relationships between AOD and CC often remain".

The referee makes a comment for the interpretation of Fig. 5, stating that "*Fig. 5 does suggest that for small bins of CC, there is no large increase in WV as AOD increases*. *However, it does not show that* 'there is no large systematic AOD retrieval bias due to aerosol swelling at increased WV'". The ref suggests that "*to address the dependence of AOD on WV (rather than WV on AOD)*" it would be helpful "*to plot a line of AOD vs WV for different WV bins*". We fail to see how such a line figure of AOD vs WV would be helpful if WV is already in bins. We wonder if this is a typos, and instead of "*different WV bins*" the referee meant "*different CC bins*". Further, if Fig. 5 does suggest that for small bins of CC, there is no large increase in WV as AOD increases, it does also suggest, viewed from a different perspective, that for small bins of CC, there is no large increase in AOD as WV increases. The referee also points out that any AOD dependence on WV would not be a bias per se. We agree with this latter point, so we rephrased the relevant parts of the text from “there is no large systematic AOD retrieval bias due to aerosol swelling at increased WV” to “there is no large systematic AOD increase at increased WV”.

Ref comment on conclusions p14015.12 and Abstract p14008.5: Taking into account the referee comment, and also our considerations (mentioned also above) about using SLP as a synoptic conditions proxy, we rephrased the respective passages to “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 co-variability, as approximated by SLP, alone”.

Specific comments

Title: We think that at this point it would not be very appropriate to include “seasonal cycles” in the title, as this would require substantial more results on seasonal cycles than what is currently presented in the manuscript or what could be included in the manuscript within its current focus, so we opt not to. We do indeed have an on-going analysis of seasonal cycles, but the results are so extensive that cannot be included here and will be presented in a future manuscript.

Abstract: a) done as suggested, b) rewritten (see above), c) rephrased (see above).

Data and methods: a) The literature is so extensive on this matter, being a research subject on its own, and it is outside the scope of the present paper to review it. The proposed references might also not be the most appropriate ones for the present paper, e.g., Varnai and Marshak (2009) examine the 3-D cloud effects on MODIS observations of clear sky reflectance, and they are not very appropriate at this point, while Huang et al. (2011) examine the effects of this cirrus on AOD, and, while interesting, we also think we should not enter into this discussion here. The two papers referenced in our work present results regarding the quality of both AOD (Remer et al., 2005) and WV retrievals (King et al., 2003). We added also a more recent paper by Remer et al. (2008). b) Indeed Grandey and Stier (2010) identified a scale problem in aerosol-cloud-interaction (ACI) studies, but as they note, “*Analysing satellite datasets over large regions may introduce spurious relationships between aerosol and cloud properties due to spatial variations in aerosol type, cloud regime* *and synoptic regime climatologies*". Also (Figs. 6 and 7 of Grandey and Stier), this problem is much less pronounced over land (where our study was conducted, being smaller than 5% for the scale sensitivity of N_e to τ_a up to $8^0\times8^0$ regions and also being smaller than 5% for the scale sensitivity of r_e to τ_a up to $8^0\times8^0$ regions. As our three studied regions range from $2^0\times3^0$ (PRD) to $4^0\times5^0$ (BTH), no significant scale problems are expected according to the Grandey and Stier results on the issue.

Results and discussion: a) "to exclude" changed to "to exclude, at least partially". b) we will also add all the relevant Terra results in the Supplementary material as Figs. S4 to S6., see below at Annex I of this response. c) this would indeed be an interesting exercise, but see our response to the Specific Comment on the manuscript title above. d) sentence amended (see above in our response to the refs major comment). e) rewritten, see our response to major comment, above. f) see above in our response to the refs major comment. g) Reference to Meskidze et al. added.

Conclusions: a) See our response to major comments, above. b) Sentence rephrased, see our response to major comments, above. c) Indeed. We added the following sentence to the relevant discussion section part (not the conclusions section), p14014.12: "Also, the results suggest a profound interference of the hydrological cycle with the aerosol climatic impact, that needs further investigation. Recent studies also point out to different aspects of the aforementioned interference (Grandey et al., 2014, Gryspeerdt et al., 2015, Rosenfeld et al., 2014)". d) Statement revised, see our response to major comments, above.

Additional references: Rosenfeld et al. (2014) ref. added (see response on conclusions c) above. Ref. to Gryspeerdt et al., GRL, (2014) added also, as Gryspeerdt et al., 2014b, and included in the Discussion section part that deals with CTP ("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.").

Figures:

Figure 1. a) Label applied in revised version of the figure. b) The MODIS TERRA AOD_{550} global average is estimated at ~0.1523 for the period 2003-2013. This value is given now in the caption. c) Y-axis label applied in embedded figure in revised version of the figure. d) Acronyms explained in caption.

Figures 2-4a), b), c): We considered making the proposed adjustments to the figures, but prefer not to implement them as we think this will deteriorate slightly the clarity of the figures (the figs. will have to get smaller).

Figure 5: Again, we considered making the proposed adjustment to the figure, but prefer not to implement it as we think this will deteriorate slightly the clarity of the figure (the fig. will have to get smaller).

Figures S1 and S2: No, these are daily data, smoothed with a 20-day moving average. This will be clarified in the caption in the revised version.

Tables S1-S5: Yes, a=0.05 means the same as p=0.05. a will be changed to p in the revised version, to avoid misunderstandings.

Technical corrections/suggestions

All corrected as suggested by the referee.

Annex I: Figs S4 to S6 to be added to the Supplementary Material:

Captions of Figs. S4 to S6.

Figure S4. MODIS TERRA, 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 at the cloud data color bar denote no values or less than 6 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 line graph CC-AOD and CTP-AOD relations were calculated by averaging CC and CTP within 0.1 AOD bins for several 1cm WV classes.

Figure S5. As in Fig. S4, but for the Yangtze River Delta (YRD) urban cluster.

Figure S6. As in Fig. S4, but for the Pearl River Delta (PRD) urban cluster.

Response to Referee #2

We thank the referee for the positive comments and the careful remarks.

Concerns

See the Tables S1 and S2 in the Supplement, containing detailed statistics of the AOD-CC relationship over each of the three urban clusters studied. A detailed seasonal analysis is underway, but the involved amount of work is such that the results, once finished, will be the subject of another manuscript. We can only state here, if the sample size within each urban cluster is of concern to the referee, that there are no large differences in the sample size. Further, as we state in the manuscript, the SLP classification results to some degree to a seasonal classification, as most SLP values less than 1008 mb are encountered during the warm period May-August, while most SLP values greater than 1017 are encountered during the cold period October-March. See also the now included in the revised version Figs. S7 and S8 of the Supplement, with the requency distribution of the AOD-CC values for different SLP levels and for different WV levels (AOD<0.6). A reference to Wang et al. (2015) has now also been added to the manuscript.

Specific comments

1 P14013 lines 13-14: This is a very interesting remark, but cannot be clarified within the present work as it involves very substantial amounts of work, that would go far beyond the scope of the present paper. As the referee correctly points out, to get an insight into this a classification of aerosols and the cloud layering pattern would be needed. This would require data from other sensors than MODIS, as MODIS does not provide information on the vertical aerosol-cloud layering pattern, nor it is possible to use MODIS for aerosol classification over land. The latter is due to the fact that for a classification (e.g. (e.g. according to Barnaba and Gobbi, 2004 or Pace et al., 2006), the Fine mode ratio (FMR) or the Angstrom Exponent would be needed, which, although provided by MODIS are not accurate over land. As we note in the manuscript, personal communication with L. Remer, NASA GSFC: "Angstrom over land is not reliable and we recommend strongly not to use it". The same exactly stands for the FMR product which gives only an indication of the existence of dust/non-dust aerosols being more a crude qualitative measure (see also Georgoulias and Kourtidis, 2011). Even if the above considerations were not valid, further classification of the dataset would result in much fewer data points within each subclass and this would deteriorate the statistics considerably. See also the now introduced Table S5 of the revised Supplement, for detailed statistics of the AOD-CC relationship for different WV classes over PRD.

2 and 3, on the use of daily SLP instead of hourly (or 6 hourly) close to overpass time: The reason for this was that firstly the amount of work would increase and secondly (and most importantly) this would not lead to any substantial improvement in the calculations or a more accurate partitioning into the SLP classes. This is because, we report here only for the two classes that correspond to the core of high and low pressure systems. Even if such a system moves quickly it would take days to pass over the area, not hours.

4 P14011 lines 12-15: More details are now provided in the revised manuscript, as follows: ”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 timeseries of concurrent AOD, WV, CC and CTP values, the data were bined in equally sized bins (thus not equal sample size bins, as this would make 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 X 10 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 X 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)”.

5 unequally sized bins: Correct, each bin of meteorological factor and AOD does not have equal sample size. Using equal sample sizes would result in different bins in the three different urban clusters studied and would make any comparison difficult. The statistical analyses performed (see Tables S1 to S5 of the revised Supplement) show that this has not affected the conclusions or introduce a bias.

6 P14012 second paragraph: Paragraph moved to Section 2 as suggested.

7 P14016: Fig. 5 does suggests clearly that there is no large increase in WV as AOD increases, hence we maintain that it does also suggest, viewed from a different perspective, that there is no large increase in AOD as WV increases, as where WV values are largest AOD values are not. We rephrased slightly the relevant parts of the text, though, from "there is no large systematic AOD retrieval bias due to aerosol swelling at increased WV" to "there is no large systematic AOD increase at increased WV".

References:

Barnaba, F. and Gobbi, G. P.: Aerosol seasonal variability over the Mediterranean region and relative impact of maritime, continental and Saharan dust particles over the basin from MODIS data in the year 2001, Atmos. Chem. Phys., 4, 2367-2391, doi:10.5194/acp-4-2367-2004, 2004.

Georgoulias, A. K. and Kourtidis, K. A.: On the aerosol weekly cycle spatiotemporal variability over Europe, Atmos. Chem. Phys., 11, 4611-4632, doi:10.5194/acp-11-4611-2011, 2011.

Pace, G., di Sarra, A., Meloni, D., Piacentino, S., and Chamard, P.: Aerosol optical properties at Lampedusa (Central Mediterranean). 1. Influence of transport and identification of different aerosol types, Atmos. Chem. Phys., 6, 697-713, doi:10.5194/acp-6-697-2006, 2006.

Annex: Newly introduced Figs. in the revised Supplement

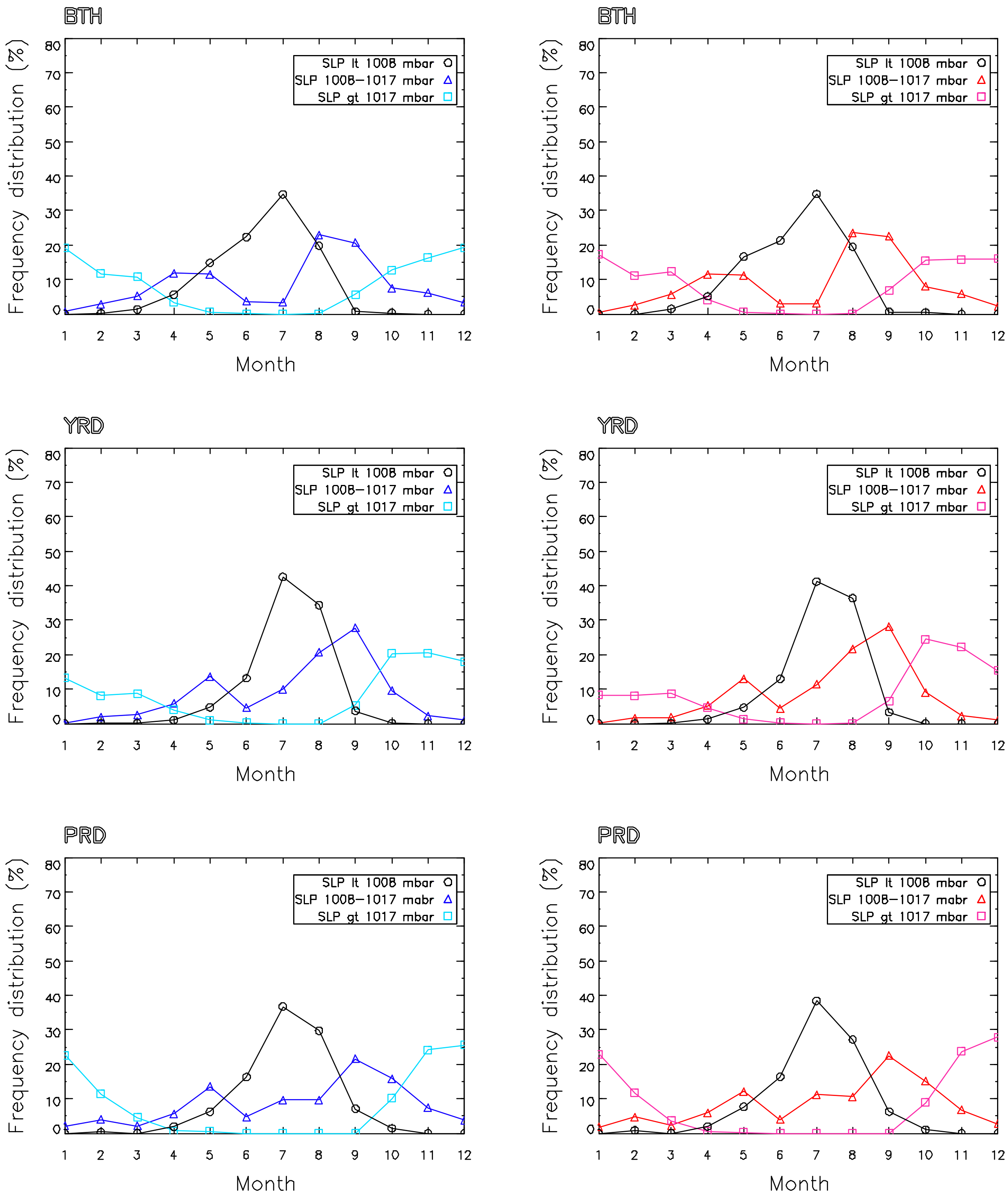

Fig. S7: Frequency distribution of the AOD-CC values for different SLP levels (AOD<0.6). Left: AQUA, right: TERRA.

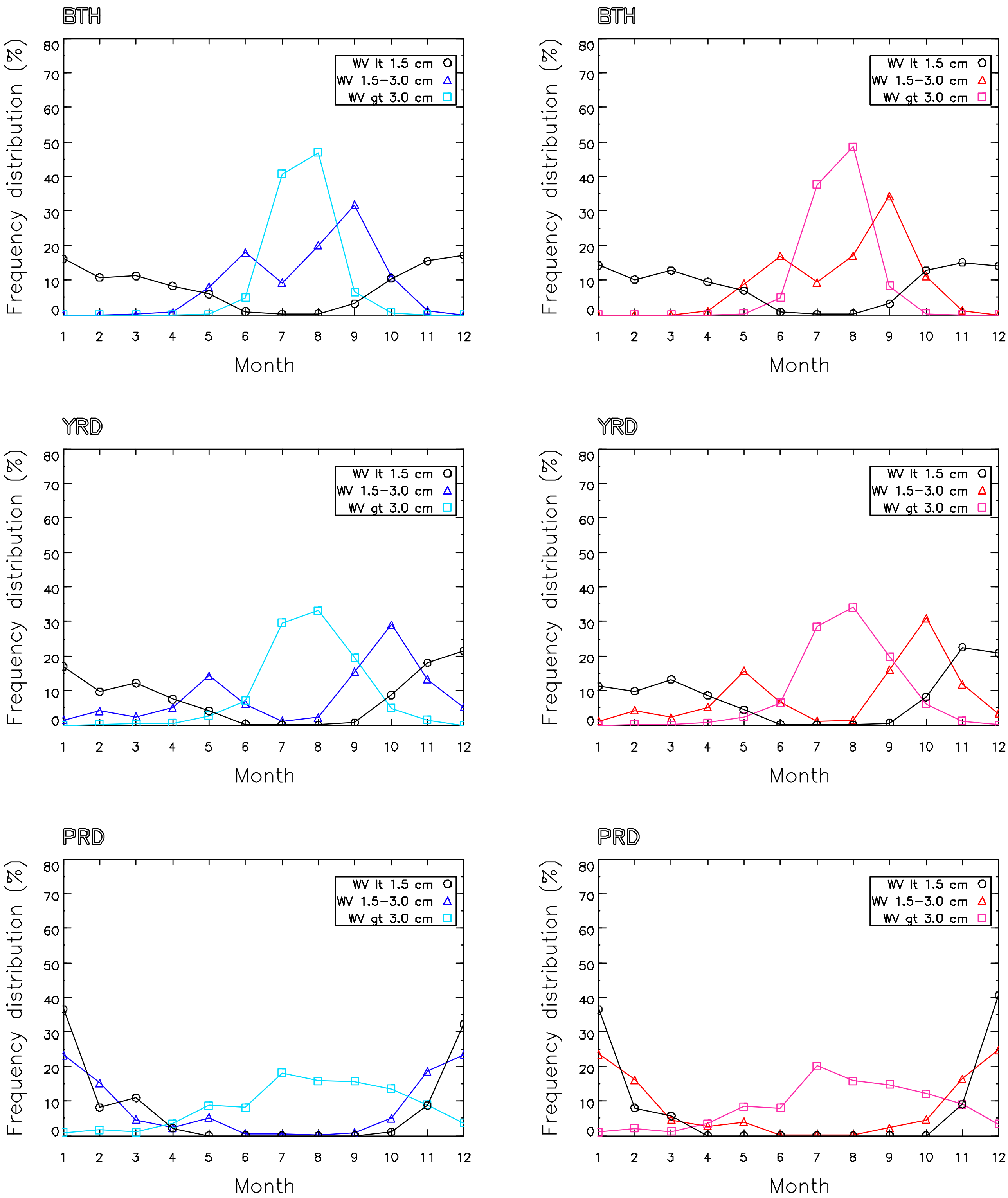

Fig. S8: Frequency distribution of the AOD-CC values for different WV levels (AOD<0.6). Left: AQUA, right: TERRA.

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, and Cloud Top Pressure (CTP)

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three

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observations

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satellite-retrieved data

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cannot be explained

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is not solely due to

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, as approximated by SLP, alone

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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.

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Hence, studies of AOD-CC relationships based on satellite data, might greatly overestimate or underestimate the AOD impact on CC in regions where AOD and WV have similar or opposite seasonal variations, respectively.

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Cloud Top Pressure (

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short-wave

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Absorption in the atmosphere, by perturbing the vertical temperature structure, may impact also clouds (semi-direct effect, Ackerman et al., 2000).

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over

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Remer et al., 2008,

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and to investigate the influence of synoptic meteorological conditions on the AOD-CC relationship (Fig. S3 in the supplementary material), and to exclude, at least partially, artifacts on the AOD-CC relationship resulting from synoptically induced co-variance (Mauger and Norris, 2007; Loeb and Schuster, 2008; Quaas et al., 2010; Gryspeerdt et al., 2014a), the AOD, CC, WV and CTP MODIS data were classified into 3 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 timeseries of concurrent AOD, WV, CC and CTP values, the data were bined in equally sized bins (thus not equal sample size bins, as this would make 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 X 10 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 X 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).

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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 etc.), and hence it is omitted from the discussion.

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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 etc.), and hence it is omitted from the discussion

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Figs. S4 to S6 in the supplementary material

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Wang et al. (2015) also noted the different humidity levels during summer and winter over East China.

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Hence, studies of AOD-CC relationships based on satellite data that do not take into account WV, 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.

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Hence, studies of AOD-CC relationships based on satellite data that do not take into account WV, might greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations.

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even after accounting for WV and synoptic variability as manifested by SLP, weakened positive relationships between AOD and CC often remain

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AOD does have an impact on CC even if synoptic and WV variability are accounted for

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Also, the results suggest a profound interference of the hydrological cycle with the aerosol climatic impact, that needs further investigation. Recent studies also point out to different aspects of the aforementioned interference (Grandey et al., 2014,

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Gryspeerdt et al., 2015, Rosenfeld et al., 2014

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Gryspeerdt et al., 2015,

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; Gryspeerdt et al., 2015

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retrieval bias due to aerosol swelling

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, Figs. S7 and S8 of the Supplementary material

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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.

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AQUA and TERRA (Meskhidze et al., 2009) are

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(compare Figs. 2 to 4 with Supplementary material

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Namely, studies of AOD-CC relationships based on satellite data, might greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations.

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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 co-variability, 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

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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.

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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 (Albrect, 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, by perturbing the vertical temperature structure, may impact also clouds (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 clusters are among the most dynamic and rapidly growing regions of China. Several such clusters have emerged in the past two 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 some times 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 vapour amounts on AOD-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 conjunction with sea level pressure (SLP) from NCEP/NCAR reanalysis data.

2 Data and Methods

The 3 major urban clusters of China have been selected so as to be representative of 3 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 (Domrös 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^o-40.5^{o}N, 113.5^o-120.5^{o}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^o-33.5^{o}N, 117.5^o-123.5^{o}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^o-24.5^{o}N, 111.5^o-115.5^{o}E] is an area within the Inter-Tropical Convergence Zone (ITCZ) migration belt, with high anthropogenic aerosol emissions (Streets et al., 2003; Streets et al., 2008; Lei et al., 2011). Over the 3 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^ox1^o 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 (AOD_{550} or just AOD), CC, WV for clear conditions (Remer et al., 2005, Remer et al., 2008, King et al., 2003) and CTP from both satellites. Aerosol index (AI), defined as the product of the AOD and Angstrom exponent, is a good proxy to quantify cloud condensation nuclei and has been applied in many previous ACI studies (e.g., Costantino and Breon, 2010 [off-coast Namibia and Angola], 2013 [over the SE Atlantic Ocean]). 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 Angstrom exponent in the derivation of AI. Namely, the Angstrom exponent is not reliable over land areas. We quote a personal communication with L. Remer, NASA GSFC: "Angstrom 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^o x 2.5^o NCEP/NCAR SLP data were regridded using bi-linear interpolation in order to match the MODIS $1^o \times 1^o$ level-3 dataset.

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 (Fig. S3 in the supplementary material), and to exclude, at least partially, artifacts on the AOD-CC relationship resulting from synoptically induced co-variance (Mauger and Norris, 2007; Loeb and Schuster, 2008; Quaas et al., 2010; Gryspeerdt et al., 2014a), the AOD, CC, WV and CTP MODIS data were classified into 3 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 timeseries of concurrent AOD, WV, CC and CTP values, the data were bined in equally sized bins (thus not equal sample size bins, as this would make 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 X 10 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 X 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).

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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 timeseries of AOD, CC, CTP and WV from MODIS TERRA and AQUA satellites over 5 grid points where cities of the 3 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 5 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 0.3-1.4, while over YRD they are between 0.5-1.3 and between 0.5-1 over the PRD urban cluster. BTH, with an average AOD550 of 0.654±0.15 during the study period (2003-2013) experiences somewhat heavier aerosol loading than the other 2 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 large trends are apparent during the study period (Figs. S1 and S2 in the supplementary material).

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To investigate the influence of synoptic meteorological conditions on the AOD-CC relationship (Fig. S3 in the Supplement), and to exclude, at least partially, artifacts on the AOD-CC relationship resulting from synoptically induced co-variance (Mauger and Norris, 2007; Loeb and Schuster, 2008; Quaas et al., 2010; Gryspeerdt et al., 2014a), the MODIS data were classified into 3 SLP classes (from NCAR/NCEP data, see above) and examined the AOD-CC-WV relationship at the low and high SLP classes.

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Figs. 2-4 show the AOD-CC-WV relationship over the 3 urban clusters studied for two of the three SLP classes, namely low SLP (SLP<1008 hPa, Figs. 2-4 a,b) and high SLP (SLP>1017 hPa, Figs. 2-4 e,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 etc.), and hence it is omitted from the discussion (fig. for 1008<SLP<1017 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 1cm bins and AOD is in 0.1 bins. The same analysis was also performed with MODIS TERRA data (Figs. S4 to S6 in the Supplement), 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,f, 3a,b,e,f and 4a,b,e,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>3cm occurs (Figs. S7 and S8 in the Supplement). 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 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 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. 2-4 a,b,e,f). In fact, over PRD the impact of AOD on CC for constant WV seems negligible (Fig. 4a,b,e,f). In the other two regions, BTH and YRD, CC might increase by up to 0.1 at 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

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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 of the supplementary material.

Hence, studies of AOD-CC relationships based on satellite data that do not take into account WV, 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 the AOD-CC relationships (e.g. Mauger and Norris, 2007; Quaas et al., 2010; Koren et al., 2010; Engstrom and Ekman, 2010; Chand et al., 2012; Grandey et al., 2013). Most importantly, it is in agreement with recent reports that gave 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. 2-4 a,b,e,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 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, etc.) observed in some regions of the planet through the human 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, that needs further investigation. Recent studies also point out to 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 (Fig. 5). This indicates that there is no large systematic AOD increase at increased WV.

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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 of the Supplement, and also compare parts b and f of Figs. 2-4).

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CTP is a cloud parameter that can be used as a proxy for cloud vertical development. Hence a number of recent studies 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 globally (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 pressure system and WV bins (Figs. 2-4 c,d,e,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. 2-4 c,d), while at high SLP it was not impacted by AOD in all three urban clusters (Figs. 2-4 e,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 Supplement 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).

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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 3 major urban clusters of China, representative of 3 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 co-variability, as approximated by SLP, alone. It is found that at AOD>0.2 the AOD impact on

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CC at low SLP conditions is about two times higher than its impact at high SLP conditions. Further, at its largest part this difference is due to WV differences between low and high SLP conditions rather than arising from differences in horizontal transport patterns. Hereupon, we stratified the data into 3 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 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.

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Διαγράφηκε: Namely, studies of AOD-CC relationships based on satellite data, might greatly overestimate the AOD impact on CC in regions where AOD and WV have similar seasonal variations, while they might greatly underestimate the AOD impact on CC in regions where AOD and WV have opposite seasonal variations.

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 doesn't seem to impact significantly CTP. 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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NCEP/NCAR reanalysis sea level pressure data (http://www.esrl.noaa.gov/psd). This research has been financed partly by EPAN II and PEP under the national action “Bilateral, multilateral and regional RandT cooperations” (AEROVIS Sino-Greek project) and partly under the FP7 Programme MarcoPolo (Grand Number 606953, Theme SPA.2013.3.2-01). A.K. Georgoulias also received funding from the European Research Council under the European Union’s Seventh Framework Programme (FP7/2007-2013)/ERC grant agreement no. 226144 (C8 project).

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Figure 1. Map of China with the ratio of local AODs to the global mean AOD (~0.1523) for the period 2003-2013. The position of the 3 urban clusters studied here (white squares, Beijing-Tianjin-Hebei: BTH, Yangtze River Delta: YRD, and Pearl River Delta: PRD) and their 5 major cities (white crosses) is marked. Estimates of the population (in millions, M) for the period 1990-2015 for the 3 urban clusters [CIESIN/CIAT, 2005] are also embedded in the map.

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Figure 2. MODIS AQUA, 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 at the cloud data color bar denote no values or less than 6 values in this bin. Figures on the left present average CC and CTP values in 1cm WV and 0.1 AOD bins while figures on the right present results as line graphs. The line graph CC-AOD and CTP-AOD relations were calculated by averaging CC and CTP within 0.1 AOD bins for several 1cm WV classes.

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

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

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 at the cloud data color bar denote no values or less than 6 values in this bin.

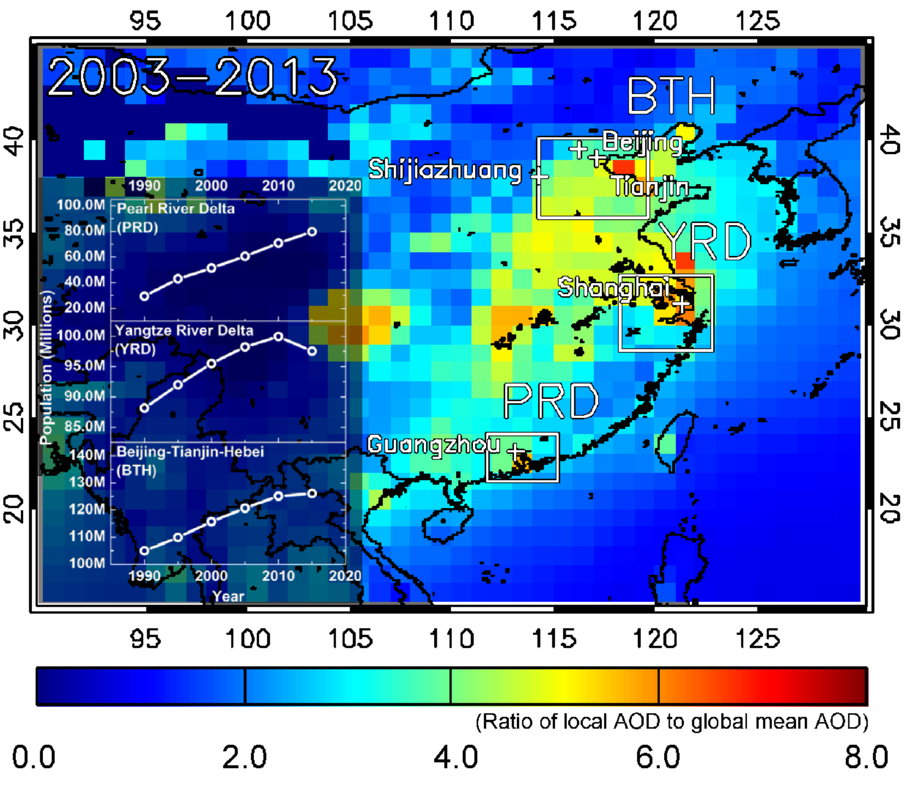

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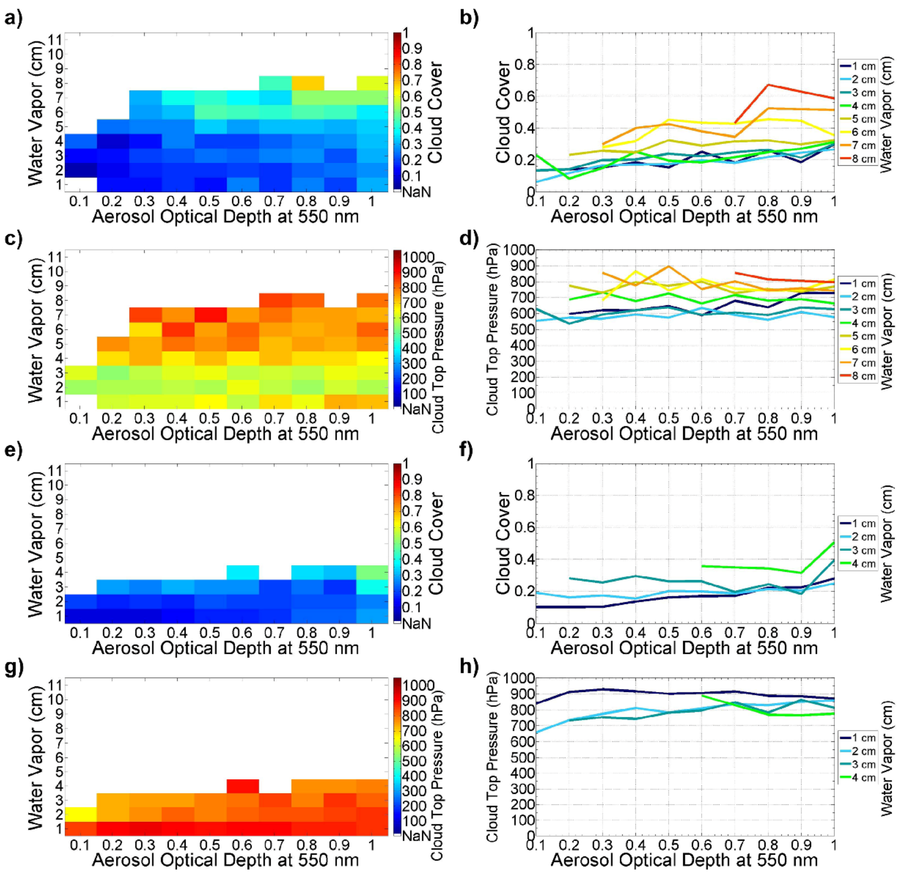
Figure 1.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Water Vapor (cm)
Aerosol Optical Depth at 550 nm
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Cloud Top Pressure (hPa)
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Figure 2.

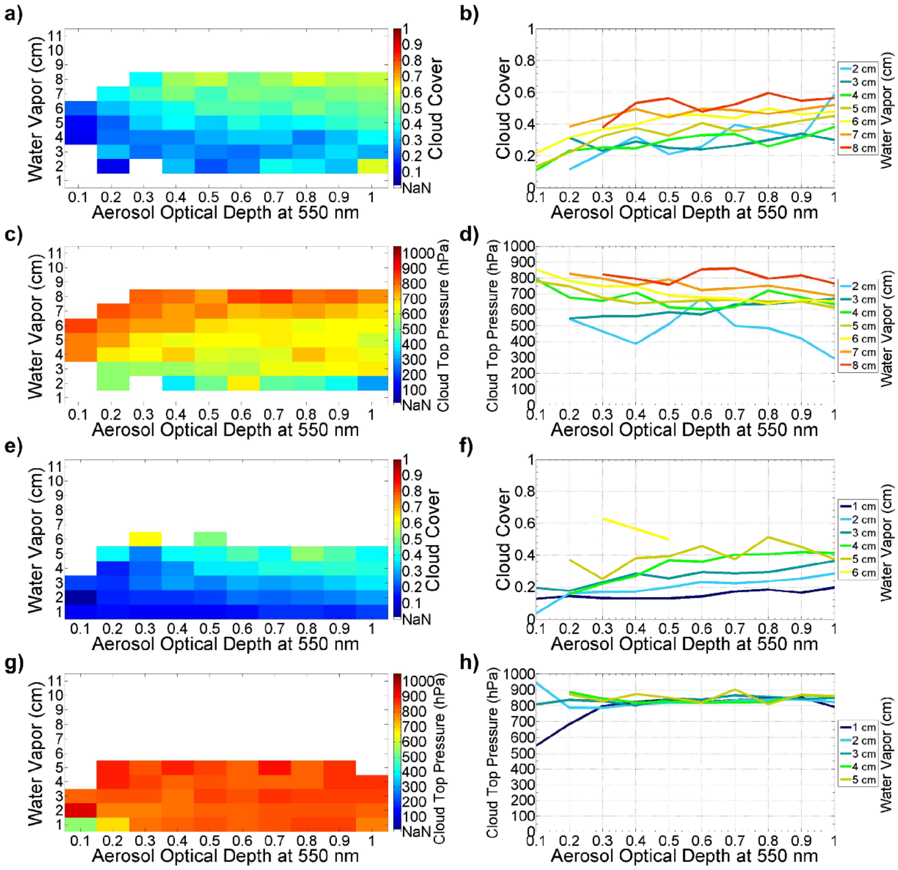
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Figure 3.

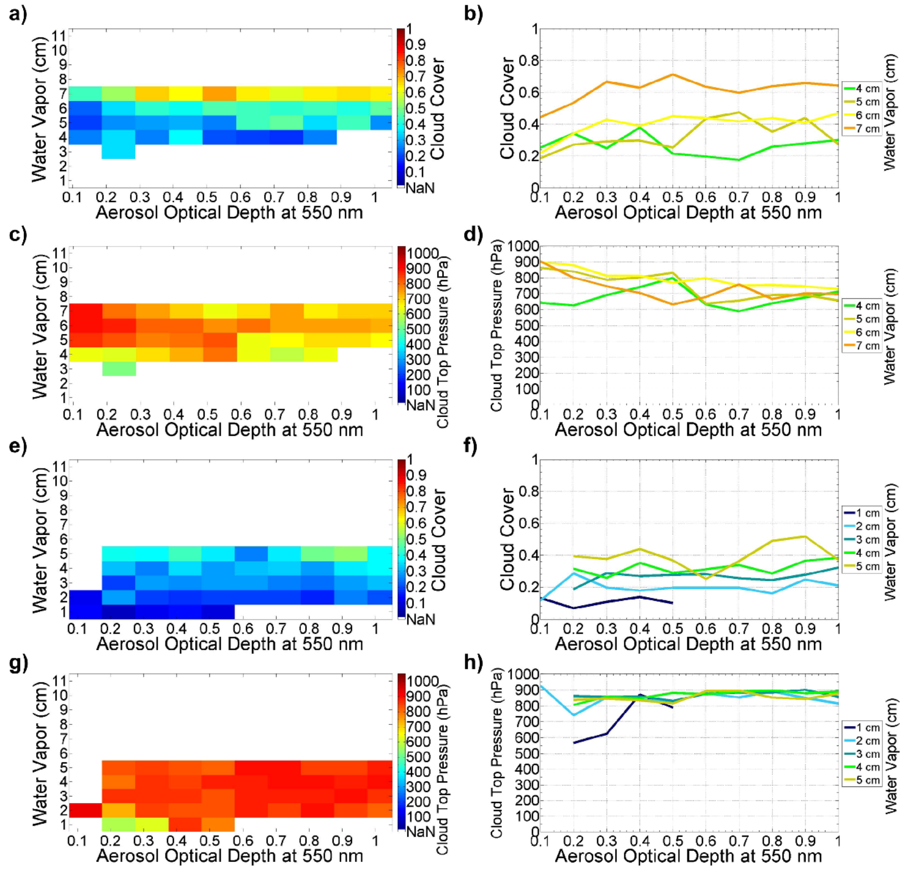
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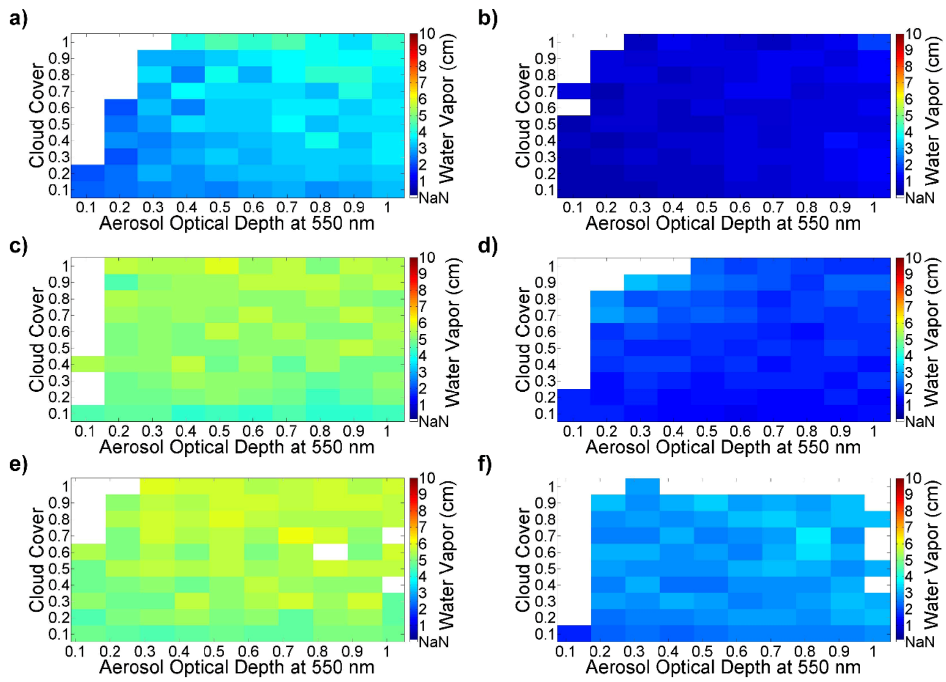
Figure 4.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Cloud Cover
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Aerosol Optical Depth at 550 nm
Water Vapor (cm)
1 2 3 4 5 6 7 8 9 10
NaN

Figure 5.