Observational evidence for aerosols increasing upper tropospheric humidity

Reply to Anonymous Referee #1

Laura Riuttanen et al.

Atmospheric Chemistry and Physics Discussions 14.10.2016

Comments by Referee #1:

General Comments:

This is a very interesting study to investigate the impacts of aerosol on the increase in upper tropospheric humidity by using remote sensing datasets over the ocean east of China. The study shows that increased aerosol loads are associated with higher upper tropospheric humidity via changes in the microphysics of deep convection. Based on long-wave radiation transfer calculation, the authors concluded that an increase in upper tropospheric humidity leads to a positive regional radiative effect. The results are well presented and structured, and the topic is suitable for publication in Atmos. Chem. Phys. after addressing some specific comments listed below.

Authors' response:

We thank the Anonymous Referee #1 for his / her comments. Please find our response to the specific comments below.

Specific Comments:

An increase in the number of atmospheric aerosols acting as cloud condensation nuclei (CCN) would slow down the diffusion growth of droplets, and thus smaller cloud droplets. To better support this mechanism using observational evidences, cloud particle effective radii (or cloud albedo) and cloud fraction, which can be retrieved by remote sensing observations, are suggested to be included in the analysis.

The referee suggests adding cloud properties by satellite instruments to the study. Cloud processes are an essential step in the proposed AOD-UTH causality chain. However, as deep convective clouds develop quite rapidly, data with a very fine time resolution would be needed. We definitely think such a study would be worthwhile, but would not be within the scope of this paper as a different source of data would be needed (e.g. from a measurement campaign).

Several studies have already shown that in polluted environments cloud droplets and ice crystals associated with deep convective clouds are smaller and more numerous than in clean environments. Both observational studies (e.g. Koren et al. 2005; Sherwood 2002; Jiang et al. 2009) and modelling studies (e.g. Fan et al. 2013; Khain et al. 2005; Morrison and Grabowski 2011; Storer and van den Heever 2013) show it (see also IPCC AR5, Boucher et al. 2013). We have added these references to the revised manuscript on Section 1.

Identification of deep convective clouds might further improve the results. MODISdetected cloud top pressure and CloudSat data would be helpful to select the cases with deep convective clouds.

Also identification of deep convective clouds separately would be useful in a cloud-scale study with a lagrangean approach. Such an approach, however, would be different than in our study. We have used a precipitation limit to separate cases with deep convection to cases without precipitation or precipitation from shallow clouds. We have added the following explanation to the revised manuscript Section 2.4:

"We use the limit of 1 mm to exclude cases without any deep convection from the study."

It should be noted and be mentioned in the article that using AOD as a measure of CCN concentration may introduce substantial uncertainties which is dependent on the aerosol type, vertical profile and hygroscopic growth.

We agree with the Referee #1 that using AOD as a measure of CCN concentration may introduce uncertainties which depend on the aerosol type, vertical profile and hygroscopic growth. We have added the following sentence to section 2.1:

"However, it should be noted that AOD does not provide information on aerosol type, vertical profile of the aerosols or hygroscopic growth of the aerosol particles." Causality questions related to the hygroscopic growth of the particles are further discussed in section 3.2.

The discussion on long-wave radiative effect is somewhat incomplete and unclear. For instance, the time period of the calculated the top-of-the-atmosphere radiative effect should be clarified, monthly mean or instantons values, in Section 2.5. Moreover, discussions on radiative effect are too short in Section 3.3. It would be better to discuss the radiative effect in detail, such as the difference between tropical moist (TM) and mid-latitude dry (MLD) conditions.

We have added text about the radiative effect and modified chapter 3.3 as following:

"Outgoing long-wave radiation is sensitive to the upper tropospheric water vapour concentration (Held and Soden, 2000). Water vapour is a greenhouse gas that mainly affects climate by absorbing outgoing longwave radiation. To balance the decrease in outgoing long-wave radiation that results from increased UTH, an increase in surface and lower atmospheric temperatures is required that acts to increase the outgoing long-wave radiation.

To quantify these effects, radiative transfer calculations (see Section 2.5) were conducted for two reference soundings of tropical moist (TM) and mid-latitude dry (MLD) air. The results show that a 1 %RH increase in the UTH of moist tropical air causes a local positive radiative effect of 0.25 W m⁻² (0.39 W m⁻² for dry air, see Table 3). In dry air the impact is larger as the fractional increase in water vapour concentration for a fixed increase in RH is larger in dry air (Held and Soden, 2000).

The observed summertime increase of 2.2 ± 1.5 %RH in UTH (5.8 ± 1.4 %RH without removal of AOD dependence on cirrus fraction) due to aerosols corresponds to the radiative effect of ± 0.4 W m⁻² (± 1.4 ± 0.3 W m⁻², interpolated from Table 3) in tropical moist air. The real radiative effect by this phenomenon may be even larger than these values, since the effect of wet scavenging may partially obscure the microphysical effects of aerosols on UTH in our study region."

Technical Corrections:

Caption of Figure 11: please check the labels of panels in the caption.

Labels of panels in Figure 11 have been corrected.

Page 6, Line 8: MSL should be defined here.

We have replaced MSL with "mean sea level" on page 6, line 8.

Figure 10: the abbreviations like omega and dir in this figure need to be specified in the caption.

Specifications of the abbreviations "omega" and "dir" has been added to the caption of Figure 10.

References:

Fan *et al.*: Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds, *PNAS*, 110, 48, 2013.

Held and Soden: Water vapor feedback and global warming. *Annu. Rev. Energy Environ.*, 2000, 25, 441-475, 2000.

Jiang *et al.*: Aerosol-CO relationship and aerosol effect on ice cloud particle size: Analyses from Aura Microwave Limb Sounder and Aqua Moderate Resolution Imaging Spectroradiometer observations. *J Geophys. Res.*, 114, D20, 2009.

Khain *et al.*: Aerosol impact on the dynamics and microphysics of deep convective clouds. *Q. J. R. Meteorol. Soc.*, 131, 611, 2639-2663, 2005.

Koren *et al.*: Aerosol invigoration and restructuring of Atlantic convective clouds. *Geophys. Res. Lett.*, 32, 14, 2005

Morrison and Grabowski: Cloud-system resolving model simulations of aerosol indirect effects on tropical deep convection and its thermodynamic environment. *Atmos. Chem. Phys.*, 11, 20, 10503-10523, 2011

Sherwood, S.: Aerosols and Ice Particle Size in Tropical Cumulonimbus. J. Clim., 15, 9, 1051-1063, 2002.

Storer and van den Heever: Microphysical processes evident in aerosol forcing of tropical deep convective clouds. *J. Atmos. Sci.*, 2013, 70, 2, 430-446, 2013.

Observational evidence for aerosols increasing upper tropospheric humidity

Reply to Anonymous Referee #2

Laura Riuttanen et al.

Atmospheric Chemistry and Physics Discussions 14.10.2016

Comments by Referee #2:

This is an interesting paper that studied the relationships between aerosol loading and upper tropospheric humidity using satellite remote sensing data. The paper is very well written and easy to follow. The authors have comprehensively explored many elements of uncertainty, however I still have some questions on the analysis. I suggest the publication of the manuscript after addressing some comments as follows.

Authors' response:

We thank the Anonymous Referee #2 for his/her comments. The reviewer raises some questions in the specific comments. Please find our response below.

Specific comments:

1. During the summer season, there might be diurnal variation of precipitations. Since the UHR change could result from the convective transport of marine boundary layer air to the upper troposphere and AODs are measured 11 hours ahead of URH, differences in diurnal variability between precipitation (UHR) and AODs are likely to contribute to the seen relationships. Suggest test it.

We agree that there might be a diurnal cycle in convection in the area. In our analysis we have used AOD data obtained at 10:30 a.m., UTH data at 9:30 p.m. and precipitation data from 8:30 a.m. to 8:30 a.m. local solar time. As the time of the observation is always the same (once per day), diurnal cycle cannot affect our results.

2. The binning approach is not totally convincing for excluding the meteorological/ synoptic effects on AOD. In addition, the value ranges of those bins are not clear. I would think more logically one should compare the 10-m wind speed and surface humidity between high AOD and low AOD scenarios and test the statistical significance of 10-m wind speed differences and surface humidity differences between the high and low AOD scenarios. Not only that AODs are influenced by humidity but also it has significant contributions from large size natural sea salt aerosols (their emission rates are wind speed dependent).

We agree that AOD depends on both low level wind speed and surface humidity. However, the effect of AOD on UTH is seen in every class of 10-m wind speed and 2-m relative humidity – except the bin with the largest surface relative humidity. Therefore, although AOD depends on those meteorological variables, AOD's effect on UTH cannot be due to, e.g., wind speed increasing AOD and UTH separately. If wind speed would affect AOD and UTH separately, and thereby cause a positive relation between AOD and UTH, then binning by wind speed would remove, or at least diminish remarkably, such a relation.

Referee #2 asked about the values of meteorological parameters of bins in Figure 10. We have now added a mean value of each meteorological parameter in the bins to Figure 10.

3. Is it possible to only use clear sky AOD (with cirrus fraction = 0 or < 0.1), given that cirrus clouds lead to biases in AOD values? Does the results change when using only clear sky AODs?

Using only clear-sky AOD would be problematic considering the nature of the phenomenon. As the effect of aerosols on UTH is expected to occur via sublimation of anvil cirrus clouds, it is expected that some cirrus clouds remain in the same 1 deg x 1 deg grid box. So the occurrence of cirrus clouds also reflects the occurrence of deep convection in the area and therefore we would not recommend studying only cases with cirrus fraction below 0.1.

However, the results for cirrus fraction smaller than 0.1 can be seen in Figure R1. UTH in general is lower, as expected, when cirrus fraction is low. Also error estimates are larger. However, with large UTH values, the effect can still be clearly seen in the data (Figure R1 b).

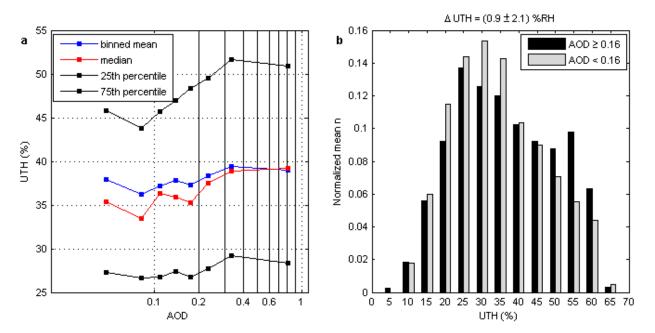


Figure R1. Same as Figure 6 in the manuscript, but only for values with cirrus fraction below 0.1. Number of data points was 4636.

Minor comments:

Page 7, Lines 10 and 15: I disagree that bulk microphysics cannot capture important aerosol effects, and only spectral bin microphysics can capture the effect. Through improved coupling between aerosols and microphysics in conjunction with maybe some improvement in the bulk microphysics (including ice nucleation scheme), there is no reason that more detailed bulk microphysics cannot simulate those effects.

We have modified the last paragraph of Section 4 in the revised manuscript as follows:

"Namely, as noted by Khain et al. (2015), the only feasible option in current general circulation models is to use bulk microphysics parameterization schemes. However, bulk microphysics schemes have trouble in producing aerosols' effect on cloud cover and cloud top height. As a result, the effect of aerosols on UTH is not correctly included in the current projections of climate change produced by general circulation models."

Observational evidence for aerosols increasing upper tropospheric humidity

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List of all changes in manuscript

(line numbers same as in the marked revised manuscript in this file)

page 2, lines 8-9: the following references and one sentence added: Morrison and Grabowski, 2011; Storer and van den Heever, 2013; Fan et al., 2013). Consistent with that, smaller ice crystals have been observed in association with increased amounts of aerosols (Sherwood, 2002; Jiang et al., 2009).

page 3, lines 5-7: the following sentence added: However, it should be noted that AOD does not provide information on aerosol type, vertical profile of the aerosols or hygroscopic growth of the aerosol particles.

page 4, line 18-19: the following sentence added: We use the limit of 1 mm to exclude cases without any deep convection from the study.

page 6, line 10: MSL replaced with "mean sea level"

page 6, lines 17-29: explanation on the radiative effect added

page 7, lines 23-30: description of the microphysics in current general circulation models modified

page 9, lines 13-14 and 29-30: two references added

page 10, lines 2 and 8-9: two references added

page 20-21: mean values of the meteorological variables added to Figure 10. That is explained in figure caption. Explanation of figure labels added to the figure caption. Units added to the figure.

page 22, lines 3-4: figure labels corrected

Observational evidence for aerosols increasing upper tropospheric humidity

Laura Riuttanen¹, Marja Bister¹, Veli-Matti Kerminen¹, Viju O. John^{2,3}, Anu-Maija Sundström^{1,*}, Miikka Dal Maso⁴, Jouni Räisänen¹, Victoria A. Sinclair¹, Risto Makkonen¹, Filippo Xausa¹, Gerrit de Leeuw^{1,5} and Markku Kulmala¹

¹Department of Physics, University of Helsinki, P.O. Box 48, FI-00014 Helsinki, Finland

²Met Office Hadley Centre, FitzRoy Road, Exeter, Devon, EX1 3PB, United Kingdom

³EUMETSAT, Eumetsat Allee 1, D-64295 Darmstadt, Germany

⁴Tampere University of Technology, P.O. Box 692, FI-33101 Tampere, Finland

0 ⁵Finnish Meteorological Institute, P.O. Box 503, FI-00101 Helsinki, Finland

Now at EMPA, Swiss Federal Laboratories for Materials Science and Technology, Überlandstrasse 129, 8600 Dübendorf, Switzerland.

Correspondence to: Laura Riuttanen (laura.riuttanen@helsinki.fi)

Abstract. Aerosol-cloud interactions are the largest source of uncertainty in the radiative forcing of the global climate. A phenomenon not included in the estimates of the total net forcing is the potential increase in upper tropospheric humidity (UTH) by anthropogenic aerosols via changes in the microphysics of deep convection. Using remote sensing data over the ocean east of China in summer, we show that increased aerosol loads are associated with an UTH increase of 2.2 ± 1.5 in units of relative humidity. We show that humidification of aerosols or other meteorological covariation is very unlikely to be the cause for this result indicating relevance for the global climate. In tropical moist air such an UTH increase leads to a regional radiative effect of 0.5 ± 0.4 W m⁻². We conclude that the effect of aerosols on UTH should be included in future studies of anthropogenic climate change and climate sensitivity.

1 Introduction

The effective globally averaged radiative forcing due anthropogenic aerosols has a best estimate of -0.9 W m⁻² but a wide uncertainty range from -1.9 to -0.1 W m⁻² (Boucher et al., 2013). This estimate combines a direct forcing of -0.35 [-0.85 to +0.15] W m⁻² due to aerosol-radiation interaction with an even less well-constrained forcing due to aerosol-cloud interaction, but still omits some potentially important mechanisms of aerosol forcing. Ramanathan et al. (2001) suggested that aerosol-induced suppression of precipitation in the updrafts of deep clouds can transport more water into the upper troposphere. But

even without changes in the total amount of precipitation, changes in cloud microphysics may affect upper tropospheric humidity (UTH, Bister and Kulmala, 2011), and thereby counteract some of the cooling effects of aerosols. High values of UTH have been observed to collocate with high values of aerosol optical depth (AOD, Kottayil and Satheesan, 2015), but here we present the first observational study to show that it is very unlikely that the increase in UTH is caused by something else than aerosols.

An increase in the number of atmospheric aerosols acting as cloud condensation nuclei (CCN) leads to more numerous and smaller cloud droplets, suppressed amount of warm rain, and enhancement of ice precipitation in deep convection (Khain et al., 2005; Morrison and Grabowski, 2011; Storer and van den Heever, 2013; Fan et al., 2013). Consistent with that, smaller ice crystals have been observed in association with increased amounts of aerosols (Sherwood, 2002; Jiang et al., 2009). Sublimation of the increased amount of hydrometeors in the upper troposphere can increase the UTH (Mechanism in Fig.1), which the Earth's long-wave radiative budget is highly sensitive to (Held and Soden, 2000; Udelhofen and Hartmann, 1995). Consistent with the proposed mechanism, more extensive and longer-lasting ice anvil clouds have been observed with larger amounts of aerosols, a sign of the increased amount of hydrometeors (Koren et al., 2005; Koren et al., 2010; Fan et al., 2013). A complicating fact is that the overall effect of aerosols on climate via deep convection may be very different from their effect on single convective clouds or precipitation events. For example, if aerosols invigorate convection causing more precipitation locally, there is compensation for this effect elsewhere or at a later time because total precipitation is strongly constrained by the atmospheric and surface energy balances (Grabowski and Morrison, 2011). Thus, a possible local increase (invigoration) or decrease (suppression) in the amount of precipitation caused by aerosols, has to be accounted for in the studies of the effect of aerosol-cloud interactions on climate (Invigoration problem in Fig. 1), as we do in this study. Note that in the following, we use the term 'invigoration' for both positive and negative changes in precipitation caused by aerosols.

In this work, we investigated the effect of aerosols on UTH by using satellite data of upper tropospheric humidity (UTH) and aerosol optical depth (AOD) in a study region east of China. We also used global data to conduct a preliminary study of the effect elsewhere. To exclude the invigoration effect from our analysis (see Fig. 1), we binned our data according to the amount of precipitation and compared UTH in cases of high and low aerosol amounts but with similar amount of precipitation. We show, for the first time, observational evidence that strongly suggests that aerosols increase the UTH.

2. Data and methods

2.1 Data

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UTH, i.e. the relative humidity with respect to water, in a layer between approximately 200 and 500 hPa, was obtained from the microwave humidity sounder (MHS; Bonsignori, 2007) onboard the MetOp-A satellite for 60° S to 60° N. The ascending node was used, crossing the equator northward at 9:30 p.m. local solar time. A microwave method developed by Buehler *et al.* (2008) enabled us to detect relative humidities also in the areas of anvil clouds.

The daily rainfall product, 3B42, from the Tropical Rainfall Measuring Mission (TRMM) multisatellite analysis (Huffman et al., 2007) was used in this study. These data cover the globe from 50° S to 50° N and are provided on a 0.25° ′ 0.25° grid. The TRMM daily rainfall product consists of satellite and in situ measurements during a 24 h time period starting at 22:30 UTC of the previous day (in our study region approximately 8:30 a.m. local solar time).

AOD has in general been considered as a moderate proxy of cloud condensation nuclei (CCN; Andreae, 2009). However, it should be noted that AOD does not provide information on aerosol type, vertical profile of the aerosols or hygroscopic growth of the aerosol particles. We used AOD from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument onboard the Terra satellite that crosses the equator southward at 10:30 a.m. local solar time (Remer et al., 2005). MOD08_D3 collection 5 level 3 global daily 1°′ 1° data "Effective Optical Depth Average Ocean Mean" at 550 nm was used in most parts of the study, and "Optical Depth Land and Ocean Mean" in global analysis. MODIS level 3 products are obtained from level 2 swaths along the flight, with 10 km resolution for AOD and uncertainty of ±0.03±0.05′ AOD over ocean. A satellite instrument cannot retrieve AOD below clouds, and in level 2 data also the pixels adjacent to a cloud are removed from the AOD product to avoid possible cloud contamination of the data. However, as the 1°′ 1° level 3 data is produced from the 10 km-resolution level 2 data, it is possible to obtain AOD and cloud properties for the same 1°′ 1° grid cell when this cell is partly cloudy (Fig. 2).

2.2 Cirrus correction

Cirrus clouds may contaminate the AOD values obtained from MODIS (Sun et al., 2011; Wollner et al., 2014; Huang et al., 2011). In our study region, MODIS AOD increased almost linearly with the increasing cirrus fraction (cf), obtained also from MOD08_D3 (Figs. 3 and 4). This may partly be due to a microphysical effect of aerosols, as more extensive cirrus anvils have been observed in association with higher AOD (Koren et al., 2005; Koren et al., 2010; Fan et al., 2013 and as shown schematically in Fig. 1a and b). However, AOD data contamination due to sub-visual cirrus is also possible (Sun et al., 2011; Wollner et al., 2014; Huang et al., 2011).

We removed data with cirrus fraction larger than 0.9 and used linear regression for cirrus fractions between 0.1 and 0.9 to remove the cirrus dependence from the AOD data to minimize the possible AOD data contamination (as explained in Fig. 4).

However, by doing so, we also removed any possible microphysical effect of aerosols on cirrus anvils and subsequently may have underestimated the aerosol effect on UTH. Therefore, we also report results without the cirrus correction in Table 1. It should be noted that the applied cirrus correction is designed to only assure that our results are not contaminated by sub-visual cirrus; it is not therefore a general method to be applied in all satellite data.

For sensitivity tests (Table 2), a linear regression for the cirrus fractions between 0.1 and 0.9 was applied with a fit of AOD = 0.1486 ′ cf + 0.2535 and root mean squared error of 0.3690 (cirrus correction version 2). The grid cells with a cirrus fraction larger than 0.9 were excluded from the analysis. For global data a similar cirrus correction as explained in Fig. 4 was applied, with a linear fit to global data of all seasons 2007-2013: AOD = 0.072 ′ cf + 0.100. The cirrus correction may not be

geographically or seasonally homogeneous, but for simplicity we applied the same correction to all grid cells in the global analysis.

2.3 Study period and area

We investigated the effect of aerosols on UTH in the maritime region east of China (120-149° N, 25-45° E). This area was chosen because of the advection of anthropogenic aerosols from the continent to this region and the frequent occurrence of convection (Fig. 5). We analysed data from days with precipitation for June, July and August in 2007-2013. Only data from the summer period were considered because during the other seasons, the advective wind speeds are much higher so that the 11-hour time difference between the AOD and UTH measurements could have resulted in these two quantities being determined from totally different air masses.

We used measurements of AOD preceding the measurements of UTH by 11 hours to better allow convection and microphysical changes, including sublimation of ice, to occur between the AOD and UTH measurements. Eleven hours may not be long enough to allow the sublimation of all ice associated with the large ice anvils produced in deep convection but a longer time lag could potentially lead to problems due to the horizontal advection of air. AOD was obtained from the Terra satellite crossing the equator southward at 10:30 a.m. local solar time and UTH from the MetOp-A satellite crossing the equator northward at 9:30 p.m. local solar time.

2.4 Data analysis

All the data were linearly interpolated to the same 1° 1° grid. The UTH data were binned according to daily precipitation in the same 1° 1° grid. The applied data were restricted to those cases when the daily precipitation was more than 1 mm. We use the limit of 1 mm to exclude cases without any deep convection from the study. For each grid point, DUTH was calculated as DUTH = median{UTH[AOD\genedian(AOD)]} - median{UTH[AOD\setmodian(AOD)]} for each precipitation bin, and an average was then calculated over the precipitation bins. Median(AOD) is the median of all AOD data in the study region for precipitation values of more than 1 mm day⁻¹. For global analysis median(AOD) is the median of AOD in each grid box. The 90% confidence interval of the mean was calculated as $\pm 1.725^{\circ}$ SD/ \sqrt{N} , where SD is the standard deviation of the N = 21 monthly median values, which were assumed to be independent of each other. For global analysis, a two-sided t-test was conducted over the monthly medians and only values significant at the 10% level are shown. A logarithmic fit was calculated for the binned mean values in Fig. 6a, excluding the first data point. The obtained fit is UTH = 45.5015 + 1.3680 $^{\circ}$ ln(AOD).

2.5 Radiative transfer calculations

The sensitivity of the radiative transfer to perturbations in the UTH was tested by using the libRadtran radiative transfer code (Mayer and Kylling, 2005). Reference soundings for the tropical atmosphere (Dunion, 2011) were used. We added 1, 2, 5 and 10 %RH to the 500-200 hPa layer of the reference soundings of tropical moist (TM) and mid-latitude dry (MLD) air. The

outgoing long-wave flux at the top of the atmosphere was integrated over the 4-50 micron wavelength band using the DISORT radiative transfer equation solver (Stamnes et al., 1988) and the correlated k-method by Fu and Liou (1992). The radiative effects were calculated only for the long-wave region, as the shortwave effect was estimated to be minor. Stratospheric adjustment was not taken into account.

5 3. Results

3.1 UTH increase associated with AOD

We found that UTH increased with increasing values of AOD for AOD higher than about 0.08 (Fig. 6a). From a logarithmic fit to these values, the UTH increase associated with the AOD increase, for example from 0.1 to 0.6, was observed to be 2.5 %RH. An UTH increase was not visible with very high amounts of precipitation (Fig. 7), presumably due to enhanced wet scavenging of aerosols. Namely, earlier strong precipitation may result in larger UTH occurring together with smaller AOD, which then masks the microphysical effect of aerosols on UTH (Quaas et al., 2010; Grandey et al, 2013). Overall, wet scavenging associated with both earlier and simultaneous precipitation masks the effect of aerosols on UTH and therefore our results may underestimate the true effect.

The median AOD over the study region was 0.18 for precipitation values of more than 1 mm day⁻¹. We selected this median to represent the limit between "high" and "low" AOD. The mean UTH difference between these two AOD categories (DUTH) was 2.2 ± 1.5 %RH (90% confidence interval, see Methods) for precipitation values of more than 1 mm day⁻¹ (Fig. 6b, see also Fig. 8). Without applying the cirrus correction, which removes part of the real microphysical effect, the corresponding UTH difference was much higher, 5.8 ± 1.4 %RH (Table 1). Without the precipitation binning, or with only a few bins, a larger mean difference in UTH between cases with "high" and "low" AOD was obtained (Fig. 9). UTH also increased less as a function of AOD in Fig. 6a when averaged over 10 precipitation bins than when the median values without precipitation binning were used. The fact that precipitation binning decreases the obtained DUTH may be at least partly due to the removal of invigoration effects of aerosols on precipitation.

We tested the sensitivity of the results by varying the limit between "high" and "low" AOD and also by averaging AOD over 5° ′5°. We also varied the time lag between AOD and UTH measurements, and the time span over which the precipitation was taken, up to 2 days. Furthermore, different aerosol products were tested. In general, a higher UTH was observed with a higher value of AOD. However, the magnitude of DUTH depended on the specific choices (Table 2).

3.2 Causality questions

Causalities in satellite studies of aerosol-cloud-interactions have been questioned because of meteorological covariation and aerosol humidification (Quaas et al., 2010; Grandey et al., 2013). For example, aerosol particles have been noted to swell in humid conditions leading to higher AOD even though the number of cloud condensation nuclei does not change (e.g. Quaas

et al., 2010; Grandey et al., 2013; Twohy et al, 2009). Such effects may then cause correlations between AOD and cloud properties.

Although precipitation binning should alleviate these problems, we repeated our analysis by binning the data according to different meteorological variables from ERA-Interim reanalysis data (Dee et al., 2011), in addition to binning it by the amount of precipitation. Surface parameters and 550 hPa vertical velocity in pressure coordinates at 00 UTC (approximately 10 a.m. local solar time) were used at 1° ′ 1° resolution (Fig. 10).

If meteorological covariation or humidification explained our results, binning by meteorological variables should significantly decrease the observed UTH difference between the high and low values of AOD. For cases with non-extreme values for meteorological variables (i.e. boxes 2 to 5 in Fig. 10), the results were practically the same as with binning only by precipitation.

With extreme values of some meteorological parameters (the lowest <u>mean sea level MSL</u> pressure, the highest 2-meter relative humidity and the largest pressure increase from the previous day), the results showed a weaker or even a negative relation between AOD and UTH. However, those values were likely associated with current or recent passage of low pressure systems, where the wet scavenging associated with significant amount of earlier precipitation may dominate the results. We conclude that meteorological covariation and humidification were not the reason for the observed association between UTH and AOD.

This strongly suggests a causal relationship between aerosols and UTH.

3.3 Effect on radiative transfer

Outgoing long-wave radiation is sensitive to the upper tropospheric water vapour concentration (Held and Soden, 2000). <u>Water vapour is a greenhouse gas that mainly affects climate by absorbing outgoing longwave radiation.</u> To balance the decrease in outgoing long-wave radiation that results from increased UTH, an increase in surface and lower atmospheric temperatures is required that acts to increase the outgoing long-wave radiation.

To quantify these effects, radiative transfer calculations (see Section 2.5) were conducted for two reference soundings of tropical moist (TM) and mid-latitude dry (MLD) air. According to our radiative transfer calculations (see Methods), The results show that a 1 %RH increase in the UTH of moist tropical air causes a local positive radiative effect of 0.25 W m⁻² (0.39 W m⁻² for dry air, see Table 3). In dry air the impact is larger as the fractional increase in water vapour concentration for a fixed increase in RH is larger in dry air (Held and Soden, 2000).

The observed summertime increase of 2.2 ± 1.5 %RH in UTH (5.8 ± 1.4 %RH without removal of AOD dependence on cirrus fraction) due to aerosols corresponds to the radiative effect of $+0.5 \pm 0.4$ W m⁻² ($+1.4 \pm 0.3$ W m⁻², interpolated from Table 3) in tropical moist air. The real radiative effect by this phenomenon may be even larger than these values, since the effect of wet scavenging may partially obscure the microphysical effects of aerosols on UTH in our study region.

4. Discussion and conclusions

Although China's outflow region is an optimal study region because of the aforementioned reasons, it is important to consider the relation between UTH and AOD globally. In the following we make a first attempt to understand the effect on global scales, but a more in-depth analysis would be a topic of an additional study. In Fig. 11, differences in UTH between the cases of "high" and "low" AOD averaged over three months are shown for the four seasons separately. The UTH difference was mostly positive, which supports our hypothesis, even though negative values were also observed in some locations, especially over remote oceans and in parts of the intertropical convergence zone. These negative values are mostly located far away from major aerosol sources and may thus be due to wet scavenging dominating the relationship between AOD and UTH over the effect of aerosols on the amount of ice in the upper troposphere and its sublimation. Similarly, where the DUTH is positive, it would be even more positive without wet scavenging. As wet scavenging overall leads to a smaller DUTH, it also decreases the statistical significance of the obtained results. Importantly, Fig. 11 indicates that our study region east of China is not anomalous, but that similar differences in UTH between "high" and "low" AOD are observed in many other regions over the globe.

A few caveats need to be taken into account when interpreting the global results (Fig. 11). First of all, in our study area, binning by other meteorological parameters proved unnecessary when the data were first binned by the amount of precipitation. However, in theory the same might not apply globally. Also, the AOD dependence on cirrus fraction may not be the same everywhere, as has been assumed in these calculations. However, changes in cloud microphysical properties tend to show logarithmic, rather than linear, dependence on the aerosol loading (Koren et al., 2008 and references therein), suggesting that the effect may also be large in cleaner areas.

Results from general circulation models show almost no changes in the relative humidity in a changing climate (Held and Soden, 2000; Soden et al., 2005). However, very few general circulation models have so far begun to include effects of aerosols on deep convective clouds (Boucher et al., 2013; Khain et al., 2015). Namely, as noted by Khain et al. (2015), the only feasible option in current general circulation models is to use bulk microphysics parameterization schemes. However, bulk microphysics schemes have trouble in producing aerosols' effect on cloud cover and cloud top height. As a result, the effect of aerosols on UTH is not correctly included in the current projections of climate change produced by general circulation models. However, it has been shown that bulk microphysics schemes cannot capture important aerosol effects on deep convection. For example, increased cloud top height and larger anvil clouds seen in observations, are not produced by bulk schemes. Only spectral bin microphysics, which is computationally too costly and not an option for current general circulation models, can produce such effects of aerosols (Khain et al., 2015). Since existing general circulation models cannot simulate aerosol effect on UTH, this phenomenon is not included in the current projections of climate change. Estimates of climate sensitivity that combine temperature observations with estimates of greenhouse gas and aerosol induced radiative forcing should also be reconsidered.

Author Contributions

L.R. and M.B. designed the study and wrote the paper. L.R. conducted the study. V.-M.K. participated in writing of the paper. V.J. processed the UTH data. A.-M.S. conducted the LibRadTran runs. All authors discussed the methods and results and commented on the manuscript.

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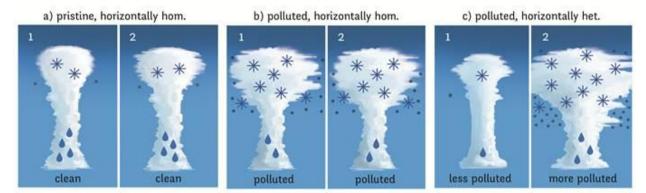


Figure 1: Deep convection in a) pristine, horizontally homogeneous, b) polluted, horizontally homogeneous and c) polluted, horizontally heterogeneous conditions. In all pairs of figures (a, b and c) the total amount of precipitation reaching the ground is the same. i) *Mechanism*. Aerosols decrease warm rain, increase ice precipitation and increase hydrometeor mass in the upper troposphere as shown in (a) and (b). More sublimation in (b) than in (a) causes an increase in the UTH in (b). *ii) Invigoration problem*. Horizontal variation of the aerosol amount, as within (c), can invigorate convection in more polluted areas (box 2 in Fig. c) and suppress convection in cleaner areas (box 1 in Fig. c). Therefore, comparing the conditions of box 2 in (c) to that in (a) would make the effects of aerosols seem too large. We avoid this problem by comparing cases with similar amounts of precipitation.

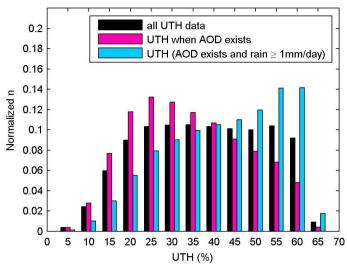


Figure 2: Number of data. Histograms of the UTH data east of China in June, July and August in 2007 - 2013. AOD cannot be retrieved in the presence of clouds, which means that we were lacking part of the data with the highest values of UTH. AOD was retrieved in approximately one third of the grid cells, in 106580 data points of the total 350407. When we limited our study to cases with precipitation ≥ 1mm day⁻¹, we had 17318 data points and the relative number of low UTH values decreased. UTH is strongly linked to precipitation, as both depend on the activity of convection.

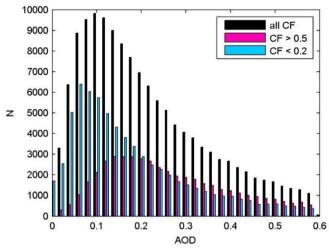


Figure 3: The relation between cirrus fraction and AOD. Histogram of MODIS AOD for all the data and segregated for small (<0.2) or large (>0.5) cirrus fraction (CF) in the same 1°′ 1° grid east of China in June, July and August in 2007 - 2013. For visual clarity, the AOD data considered here are restricted to values smaller than 0.6. The difference between these two distributions indicates that cirrus may contaminate the AOD data retrieval. Therefore, we removed the cirrus dependence from the AOD data (as explained in Fig. 4) to ensure that the relationship between AOD and UTH is not due to data contamination.

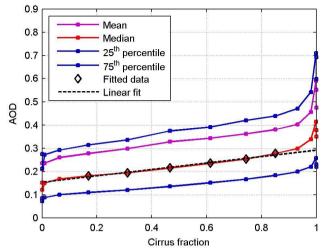


Figure 4: Dependence of AOD on the cirrus fraction. MODIS AOD at 550 nm as a function of cirrus fraction (cf) in the same 1° (1° grid east of China in June, July and August in 2007 - 2013. The dashed line shows a linear fit (AOD = 0.139 ' cf + 0.153) obtained using the median data. In order to eliminate any possible AOD data contamination by cirrus, all AOD values for the cirrus fraction between 0.1 and 0.9 were adjusted (AOD') based on the following linear fit: AOD' = AOD - 0.139 ' cf + 0.014. No correction was applied to the data for the cirrus fraction smaller than 0.1, and the data with cirrus fraction larger than 0.9 were not used in this study.

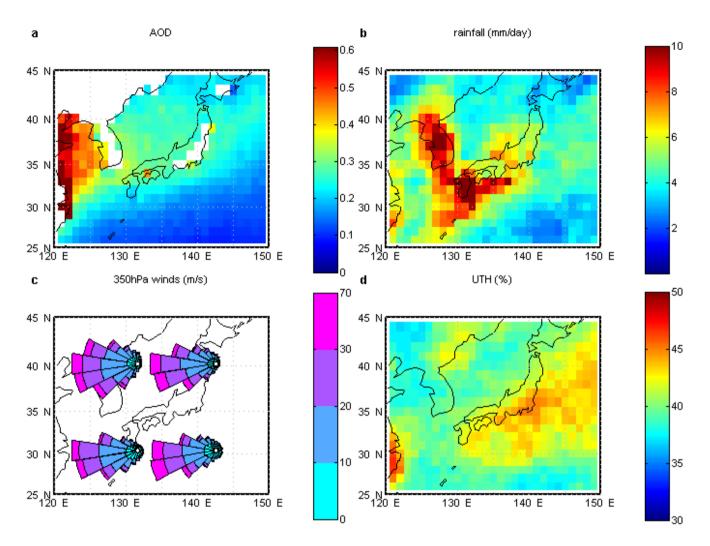


Figure 5: The study region and the data used. (a) AOD, (b) daily precipitation, (c) 350 hPa wind roses and (d) UTH in the study region east of China averaged from the observations in June, July and August in 2007 - 2013. a, Aerosol outflow from continental China clearly dominates AOD in the region, with contributions from Japan and Korea. b, Precipitation is plentiful in the region. c, Mean wind direction in the region is westerly. d, There are high UTH values downwind of the regions with large values of precipitation.

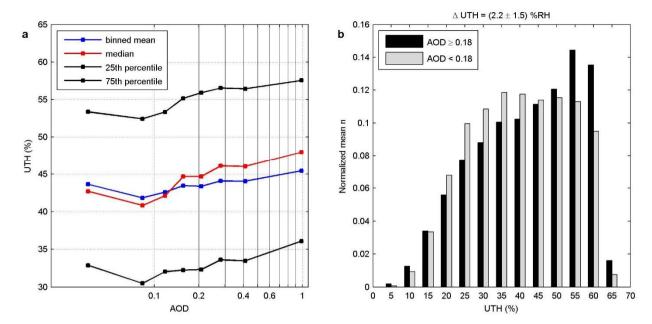


Figure 6: a, UTH as a function of AOD and b, histogram of UTH in case of low and high AOD east of China (120-149° E, 25-45° N) in June, July and August in 2007 - 2013. In both figures only data from 1°′ 1° grid cells with precipitation values of more than 1 mm / day are shown. Binned mean values are averages over 10 precipitation bins. Cirrus dependence has been removed from the AOD data (see Fig. 4).

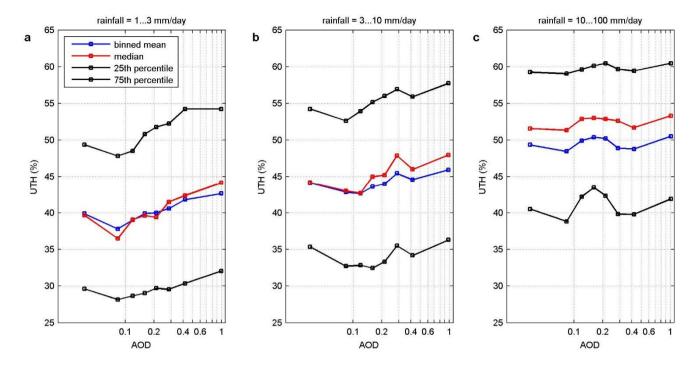


Figure 7: Results in three precipitation categories. UTH as a function of AOD (on a logarithmic scale) in three precipitation categories: (a) 1-3 mm day⁻¹, (b) 3-10 mm day⁻¹ and (c) 10-100 mm day⁻¹, east of China in June, July and August in 2007 - 2013. In each precipitation category, the AOD data were split into 8 bins, after which the 25th, 50th and 75th percentiles of the data and mean UTH of 10 precipitation bins were calculated. The cirrus dependence has been removed from the AOD data.

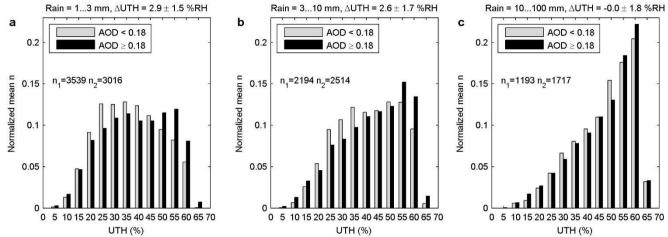


Figure 8: UTH differences in three precipitation categories. Histograms of UTH in case of "low" and "high" AOD east of China in June, July and August in 2007 - 2013, for three categories of daily precipitation: (a) 1-3 mm, (b) 3-10 mm and (c) 10-100 mm. The y-axis shows the normalized mean histogram of UTH, which is the mean of 10 precipitation bins in each precipitation category. DUTH was calculated as mean of {median[UTH(AOD≥0.18)]-median[UTH(AOD<0.18)]}, while its 90% confidence interval was calculated from the standard error of the monthly mean. The values are presented as percentage units. n1 and n2 show the number of data points in "low" and "high" AOD cases, respectively. The cirrus dependence has been removed from the AOD data.

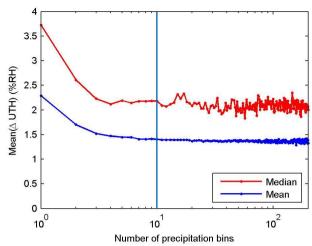
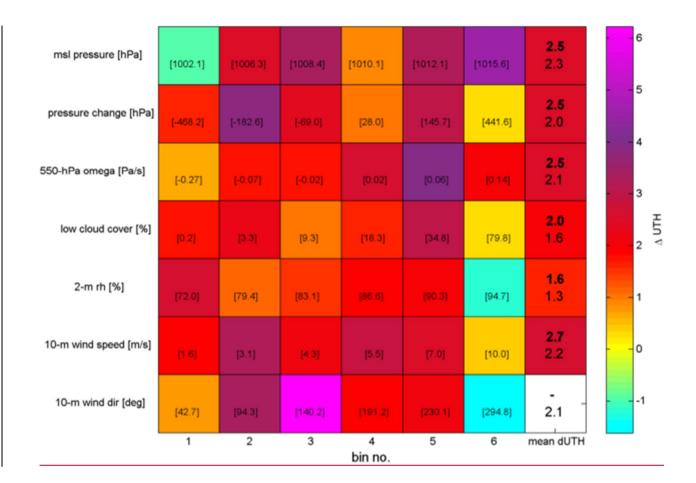


Figure 9: Effect of precipitation binning. Change in UTH (DUTH = UTH(AOD≥0.18) - UTH(AOD<0.18)) as a function of the number of precipitation bins over which the average is calculated for the data east of China in June, July and August in 2007 - 2013. Here, only the precipitation values of more than 1 mm day⁻¹ were taken into account. The cirrus dependence has been removed from the AOD data. When no precipitation binning was applied, or when only a few bins were used, the increase in UTH was clearly larger than when at least 10 bins were used. This indicates that the precipitation binning is essential if we want to remove the effect of convective invigoration on UTH. In this study, we used 10 precipitation bins (indicated by the vertical line in the figure) in order to remove the influence of invigoration on our results.



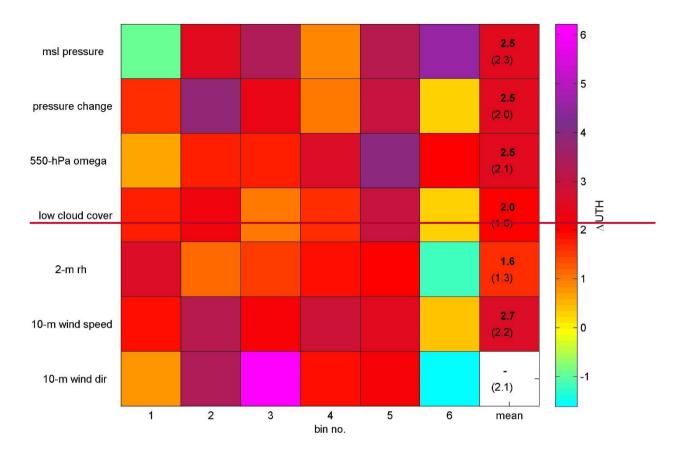


Figure 10: Meteorological covariation. After binning with precipitation, data was binned to six bins according to the value of several meteorological variables from ERA-Interim reanalysis: mean sea level (msl) pressure, pressure change from the previous day, 550-hPa vertical velocity in pressure coordinates (omega), low cloud cover, 2-m relative humidity, 10-m wind speed and 10-m wind direction (wind dir). These variables were chosen as they are expected to change considerably during the passage of synoptic (and mesoscale) disturbances. Change in UTH (DUTH = UTH(AOD \geq 0.18) - UTH(AOD <0.18)) is shown separately for the six bins and the mean over the six bins in the last column. For each meteorological variable, each of the six bins contains equal amount of data. The values increase from left to right except for 10-m wind direction for which the values go from north-easterly to north-westerly wind directions. Mean values of the meteorological variables in each bin are presented in parenthesis. In the last column, the upper value is the mean of dUTH in bins 2-5 and the lower value is the mean of dUTH in all six bins is shown in parenthesis.

If the observed increase in UTH would be caused by some of these meteorological variables, DUTH should go to zero when binned according to this variable. As no such behaviour is observed, we conclude that meteorological covariation is not the cause of the obtained results. Also, DUTH shows no systematic behaviour according to any of the meteorological variables.

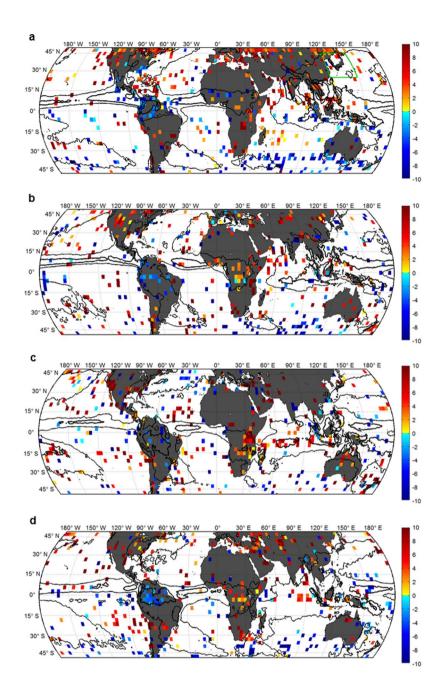


Figure 11: Mean difference in UTH (in %RH) between cases when AOD is higher and cases when AOD is lower than each grid box's median for four seasons in 2007 – 2013: (a), June, July, August, (ba) September, October and November, (cb) December, January and February, (de) March, April and May. Precipitation binning and cirrus correction were applied to the data and only grid cells with precipitation more than 1 mm / day were used. The data are averaged over 3°′ 3° grid and only grid cells with statistically significant differences (t-test with the 10% significance level) are shown. Black contours represent mean daily precipitation of 3 mm, 9 mm and 30 mm. The study region east of China is marked with a green box in (a). Wet scavenging is likely to decrease DUTH leading also to smaller statistical significance.

Table 1: Impact of different choices on the results. DUTH was calculated as DUTH=UTH[AOD≥median(AOD)]-UTH[AOD<median(AOD)] east of China in June, July and August in 2007 - 2013 and is expressed as %-units. 90% confidence interval is calculated from the standard error of the monthly mean DUTH.

	No cirrus correction	Dependence on cirrus fraction removed from the AOD data
DUTH	5.6 ± 0.9	1.9 ± 0.8
DUTH, for precipitation > 1 mm day ⁻¹	7.3 ± 1.7	3.6 ± 1.8
DUTH, for precipitation > 1 mm day ⁻¹ ,	5.8 ± 1.4	2.2 ± 1.5
precipitation binning applied with 10 precipitation bins		

Table 2: Results from the sensitivity tests. DUTH is calculated for different MODIS aerosol products, for UTH from different overpassing times and satellites, and for different precipitation measurement time spans. Small DUTH values are obtained when there is precipitation on the previous day (i.e. wet scavenging) or when UTH is measured before AOD (causality).

AOD	UTH	precipitation	DUTH	err	note
AOD ₅₅₀	MetOp-Aasc	TRMM daily	2.16	1.50	
AOD550, 0.2	MetOp-Aasc	TRMM daily	2.30	1.28	AOD limit 0.20
AOD550, ci2	MetOp-Aasc	TRMM daily	2.00	1.45	cirrus correction version 2
AOD _{land&ocean}	MetOp-Aasc	TRMM daily	2.02	1.05	same cirrus correction as for AOD 550
AI	MetOp-Aasc	TRMM daily	1.94	1.59	AI = AOD 550 * Ångström exponent
AOD_{550}	MetOp-A _{desc}	TRMM daily	0.77	0.81	UTH measured 1h before AOD
AOD_{550}	NOAA-18 _{asc}	TRMM daily	1.18	1.13	UTH measured 9h before AOD
AOD550	NOAA-18 _{desc}	TRMM daily TRMM daily +	2.42	1.26	UTH measured 3h after AOD
AOD550	MetOp-Aasc	previous day TRMM daily +	0.75	0.64	
AOD_{550}	MetOp-Aasc	following day	1.99	0.81	

Table 3: Radiative effect related to DUTH. Outgoing longwave radiation (OLR) and radiative effect (RE) caused by a decrease in OLR when relative humidity in the upper troposphere (500-200 hPa) is increased by 1, 2, 5, and 10 percentage units in the tropical moist (TM) and mid-latitude dry (MLD) air reference soundings of Dunion (2011).

	TM		MLD		
	OLR (W m ⁻²)	RE (W m ⁻²)	OLR (W m ⁻²)	RE (W m ⁻²)	
Dunion (2011)	256.83	0	269.07	0	
+1 %RH	256.58	0.25	268.68	0.39	
+2 %RH	256.33	0.50	268.30	0.77	
+5 % RH	255.61	1.22	267.21	1.86	
+10 %RH	254.46	2.37	265.55	3.52	