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Discussions

Interactive comment on "Effects of wintertime polluted aerosol on cloud over the Yangtze River Delta: case study" *by* Chen Xu et al.

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Dear editors,

The authors thank referee 1 for the prompt, thoughtful, and constructive comments (RC1). Firstly, we are sorry for our poorly written manuscript, we have asked for help from a native speaker for the revised manuscript. There is a response to RC1 review of our manuscript "Effects of wintertime polluted aerosol on cloud over the Yangtze River Delta: case study" (ACP-2016-968). According to the reviewer's suggestions, we make revision to the manuscript in detail, all of revision have been marked in red in the new manuscript (see supplement). The following is a response to comments one by one.

Question1: It seems that authors simply pair the AOD and cloud properties at a coarse grid box of MODIS measurements and perform statistical analyses without controlling

any meteorological factors. A more rigorous approach should be to carefully control the distance between the aerosol and cloud pixels to minimize the potential contamination in the retrieval. The aerosol-cloud relationships should be obtained under some controlled meteorological conditions. Answer: On our study, we performed an analysis on four sub-regions (A-D) that are located close to each other. We considered the meteorological conditions are similar between them. For the study of aerosol-cloud interactions, the most likely influencing factor regarding meteorological conditions is atmospheric humidity (water vapour). Although we did not control for meteorological conditions in the first part of our analysis, we took liquid water path into account when analysing the relationships between cloud parameters and AOD. Of course, we can not say that other meteorological parameters had no influence on this interaction (for example pressure could also have an effect – as high or low pressure systems relate to atmospheric stability). Regarding the problem of aerosol and cloud pixel matching, we have now added a case study as a part of our manuscript. In this case study, we attempt to match the corresponding parameters between aerosol and cloud over a series of days. Combined with a relatively comprehensive analysis of meteorological conditions, such as the movement of air mass, sea level pressure and so on, we attempt to use this detailed small case to inform the wider understanding of the overall analysis.

Question2: Several key technical details are described in a confusing way. For example, the manuscripts stated "AOD and cloud properties from CERESSYN" in several occasions. However, CERES doesn't directly measure aerosols and clouds. Those are from the MODIS measurements on board of Terra and Aqua. Those instrument-data relationships should be clarified in the "Data and Methods" section. A table with that information can be helpful. Answer: Thank you for your advice. It is really a good idea. For this, we collected all the information about the data sources which we used in this study, to make a table like 'Table1' in adding figure part.

Question3: What causes the different behaviors of aerosol effects over the four subregions (they are not far away in geospace)? Answer: In section 3.1, we described the difference between four sub-regions. Each sub-region has their own characteristics. Although their meteorological conditions are not too different, there are clear differences in terrain characteristics. We see more aerosol in plains and valleys with densely populated and industrialized locations, while less aerosol is found in regions that are mainly hills and mountains. The Tianmu and Dabie Mountain hinder the transport of surface contaminants away from their source regions, whilst also preventing long-distance transportation of dust from the north and marine aerosol from the east. For hilly areas with trees (region D), the surface of land has different properties from others, it may greatly effect aerosol radiation and the progress of hygroscopic growth (e.g. due to humidity levels enhanced by the trees, aerosol precursors from biomass). The terrain characteristics will easily influence the transport of pollution and affect the aerosol characteristics. The effect of hygroscopic growth depends on what is the dominating aerosol type. In addition, the CCN are governed by aerosols optical properties which depend on the aerosol chemical composition, particle size distribution and ability for particle hygroscopic growth. Thus the physical interactions between aerosol and cloud are distinct depending on the aerosol type which is linked to the regions/terrain characteristics. Therefore, we considered that the different distributions of geography and industry on this four sub-regions lead to aerosols having different dominating types and concentration ranges. These cause the different behaviours of aerosol effects.

Question4: What are the possible mechanisms to explain the non-monotonic responses of cloud properties to aerosol perturbations as shown in Figs 2-4? Answer: Cloud optical thickness (COT), Liquid water path (LWP), Cloud droplet radius (CDR) are the important parameters that reflect cloud properties. Also, their relationships with Aerosol optical depth are complex. There are many factors that affect cloud properties, like the types of aerosol, meteorological situations (G. Myhre et al., 2007) and a variety of geographical locations and seasons (Savane et al., 2015). In our study, on the same season (winter) and over a small sub-region, we consider how the amount of humidity and the dominant aerosol type affect cloud properties. At low AOD (below 0.4), increases in cloud cover are indicative of physical aerosol-cloud interactions. At

larger AOD (0.4 0.6), the increasing in cloud cover can be explained by larger hygroscopic growth near clouds (G. Myhre et al., 2007). In this range (0 0.6), the addition of aerosol causes a decrease in drop size (CDR), precipitation is suppressed, and clouds develop further (increasing of COT) before raining out (if they ever do) and last longer in the more developed stage, thus increasing the average LWP (Albrecht et al., 1989; Ferek et al., 2000). When AOD grows larger than 0.6 the cloud development is reduced, probably due to the following reasons: (a) Aerosols shade the surface, reducing surface heating and evapotranspiration so that LWP is reduced (Koren et al., 2004). (b) Absorbing aerosols (such as smoke or dust) can heat the upper levels of the troposphere, which in combination with surface shading stabilizes the atmospheric column and reduces cloud development (Koren et al., 2004,2005; Taubman et al., 2004; Ackerman et al., 2000). (c) As an increase in CCN leads to smaller droplets, evaporation around the sides and top of clouds due to mixing will become more effective at reducing the LWP (Koren et al., 2004; Burnet et al., 2007). (d) Meteorology effects, such as high-pressure systems, can inhibit convective activity, simultaneously reducing cloudiness while not allowing aerosols (from sources in the region) to “vent” away from the source region (Sinclair et al., 2010). Thus, the non-monotonic responses of cloud properties to aerosol perturbations as shown in Figs 2-4 may be explained by several of the reasons we describe above. Moreover, CDR is too complex to be analysed only with AOD. Therefore, we discuss this further in section 3.2.3 describing the various factors that can influence cloud properties for a clearer understanding.

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Question5: How to differentiate aerosol radiative and microphysical effects? Answer: Both aerosol radiative and microphysical effects are aerosol effects on climate. Aerosols lead to a number of radiative effects in the atmosphere and therefore influence the climate system, via scattering and absorption of shortwave and long wave radiation (McCormick and Ludwig, 1967). Absorption of radiation by aerosols also exerts the so-called semi-direct effect, altering the average cloud fraction by locally heating cloud- and near-cloud air layers (Hansen et al., 1997; Albrecht, 1989; Stevens and Feingold, 2009). It enhances the static stability of the atmosphere in relation to the

earth's surface and the atmospheric stratification, and may result in the evaporation of cloud droplets. The microphysical effect lies in the process of cloud formation including cloud change and dissipation. This effect is termed aerosol indirect effect (e.g. Twomey, 1974; Lohmann and Feichter, 2005). This is especially true for the indirect effect of aerosols which occurs because aerosols act as cloud Condensation nuclei (CCN). Aerosol indirect effect falls into two types: (a) Cloud albedo effect (Lohmann and Feicheter, 2005). It refers to the phenomenon of rising cloud particulate concentration and decreasing cloud radius as a consequence of the increase in aerosols that thereby alter the radiative properties of clouds, which leads to a higher albedo for clouds. This effect is also known as the first indirect effect, or the Twomey effect (Twomey, 1977). (b) Cloud lifetime (Lohmann and Feichter, 2005). This effect is a change in cloud drop number concentration which further produces change in LWP, cloud amount and radiative properties. An increase in aerosols induces a reduction in cloud particulate size and an adjustment in cloud liquid water content and cloud thickness, which reduces precipitation efficiency but prolongs cloud lifetime. From the knowledge above, it is obvious that these two effects are difficult to separate (although their effect mechanisms are different). We can't say which effect will happen solely at one time. For instance, absorption aerosol could cause a radiative effect, at the same time (if conditions are favourable), it also cause microphysical effect via acting as CCN. Over recent years the radiative effect of aerosols has been extensively studied in the literature (Chand et al., 2009; Loeb, N. G., Kato, S. 2002; Loeb, N. G., Manalo-Smith, N. 2005; Myhre et al., 2007). In our study, we focus on the process of aerosol microphysical effects on clouds, which is a different to the aerosol radiative effect.

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Question 6. What are the special aspects of aerosol-cloud interactions in winter of the YRD compared to other seasons and other regions? Answer: There has been no previous research published on the effect of aerosol on cloud during this heavy pollution period with different underlying surfaces over the YRD in winter. What we want to know is whether the high aerosol loading can induce different degrees of effect on cloud development. Wang et al. (2014) finds that CDR increases with increasing aerosol abundance over YRD during summer time. However, the characters of meteorological

conditions in winter time are different to summer, in winter being relatively static, higher aerosol loading and lower humidity. Also, the wind direction of monsoon is different from summer. It will cause differences in aerosol sources advected into/away from the region, thus influence the aerosols and their effects on cloud Tao et al. (2013) found that the different optical properties of dust-like haze clouds and notable increase in coarse mode aerosols over East China during summer agricultural burning season. Compared to more homogeneous ocean or desert areas, there are many different types of aerosols in YRD. That makes the effect on clouds more complex. Space observations show that the atmospheric aerosol load in this region is considerably higher than in the urbanized regions of Europe and North America. This is why it is an interesting region to study to research how the high aerosol load effects clouds. The results are helpful to in-depth understanding of aerosol indirect effects in Asia in fast-growing polluted areas.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-968/acp-2016-968-AC1-supplement.pdf

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Table1. Details of parameters which are used in our study.

Parameters	Products	Algorithm & Source	Satellites Channel		Resolution
AOD, FMF	CERES-SYN Edition 3A 3-hour	MODIS-derived (MOD04)	Terra and Aqua	0.55μm	1°×1° (horizontal)
COT, LWP,CTP, CLF, CDR		MODIS-Geostationary (3-hour)-derived		3.7μm (mid-IR)	
Aerosol layer fraction	CAL_LID_L2_05kmAPro-Prov-V3-30	CALIOP lidar-GMAO	CALIPSO		5km (horizontal) 60m (Vertical)
cloud layer fraction					
Aerosol vertical feature mask	CAL_LID_L2_VFM-ValStage1-V3-30				5km (horizontal) 30m (Vertical)
SLI, SLP, precipitation rate	National Center for Environmental Prediction (NCEP) Reanalysis				2.5°×2.5° (horizontal)
Air mass trajectories	HYSPLIT model				Every 6 hours at 9 key sites
$PM_{2.5}$ concentration		Air quality network in China			Daily average

Fig. 1. Table 1. Details of parameters, which are used in our study.