Dear Editor,

On behalf of all co-authors, and with their consensus we prepared the revised version of our manuscript egusphere-2023-1615 “Opposite effects of aerosols and meteorological parameters on warm clouds in two contrasting regions over eastern China” by Yuqin Liu, Tao Lin, Jiahua Zhang, Fu Wang, Yiyi Huang, Xian Wu, Hong Ye, Guoqin Zhang, Lamei Shi, and Gerrit de Leeuw. We have made all revision using track changes and also provide a clean version, in response to the comments by the three anonymous Referees. In the response files we provide a detailed point-by-point response to each Referee, including the lines numbers in the revised version where the corresponding changes have been made and a quote of these changes.

We further note that we have moved the original Section 4.4 behind the original Sections 4.5 and 4.6, to first discuss the sensitivities and adjustments of cloud parameters to changes in aerosol and confounding meteorological factors and then discuss the application of the Geographical Detector Method (GDM) to further analyze different factors influencing aerosol-cloud interaction (aci) and the effect of interactions between different factors. We also notice that the GDM provides a method to identify confounding effects.

In addition, we also introduced Section 5, Discussion, and Section 6, Conclusions.

In several Sections we aimed to better explain the novelty of our results and what is different from earlier publications on aci. In particular we aimed to stronger emphasize the use of the GDM as a complementary method to determine the effects of different factors on aci.

We thank you, as Editor, and the three Referees for the many constructive comments and helpful suggestions which helped us tremendously with improving the manuscript. In particular changing the nomenclature required a somewhat different way of thinking.

We are expecting that the three Referees are satisfied with our clarifications and ensuing revision, hopefully leading to the publication of our manuscript in ACP.

Sincerely yours,

Yuqin Liu, on behalf of all co-authors

Response to Referee #1

The authors look at the primarily at the controls on the relationship between AOD and cloud effective radius (CER), both meteorological parameters and other cloud properties. Concentrating on two regions (over land and ocean near China), this study also introduces the geographical detector method (GDM) to the study of aerosol-cloud interactions. They also look at the impact of meteorological properties on the relationship between AOD and cloud properties more generally.

The introduction of the GDM to this study is novel and of interest to the readers of ACP. With some extra explanation, I think this paper would make a really useful introduction to the method for others in atmospheric science. However, I have a number of concerns about the paper as a whole that would have to be addressed before I would recommend publication.

The authors are grateful to Referee #1 for the valuable time spent on thorough reading our manuscript and providing expert views to guide us for improving the manuscript with the main and specific points and the references. We have taken notice of all comments, listed below in black, and made many changes to the manuscript to address these, together with the comments from the other referees. We address each of your comments below and refer to our responses in the revised manuscript and provide line numbers and copy text in "quotes".

To ensure that the data used only included single layer liquid clouds and nonprecipitating cases, the filtering criteria described by Saponaro et al. (2017) were applied. It is noted that all the figures have been updated throughout the revised manuscript.

Main points

1. The introduction of the GDM method is a really nice aspect of this study. However, I feel it could be explained and examined in more detail, as there are a number of factors that are unclear to someone meeting this method for the first time. For example, the impact of the explanatory variables in Tables 4 and 5 sum to over 100%. This is not what I would have expected. Similarly, I am not familiar with the 'interaction detector' or the 'interactive q-values'. What do these mean? How should they be interpreted? Likewise, the term 'nonlinear enhancement of the influence of the independent parameters' on L456 is not straightforward to someone new to this method.

Answer: In statistics, the q-value is a measure used to evaluate the explanatory power of variables on the dependent variable. When multiple independent variables are considered separately, it is indeed possible for the sum of the q-values of multiple X variables to exceed 100%. When they are considered together, this is referred to as 'interaction q-value'. This situation is quite common and similar to the issue in multiple linear regression. The main reason for this is the presence of correlation among the X variables, indicating that these variables are not independent. Consequently, multiple independent variables may contribute to the dependent variable in a similar manner, leading to a sum of q-values over 100%.

To better explain this and clarify "interaction detector" and "interaction q-values", we have replaced the text below figure 2 (lines 354-373) with "The interaction detector can be used to test for the influence of interaction between different influencing factors, e.g., x1 and x2, on the dependent factor (y) and whether this interaction weakens or enhances the influence of each of x1 or x2 on the dependent variable, y, or whether they are independent in influencing y. For example, Figure 3(a) shows the spatial distribution of the dependent variable, y. The factors x1 and x2 both vary across the study region, but in different ways, and for each factor different sub-regions can be distinguished by application of the Jenks classification method described above to each factor separately. This is illustrated in Figures 3(b) and 3(c) where, as an example, three different sub-regions are considered for each factor. Usually, the dependent variable y is influenced by several different factors xi (Figure 3) and the combined effect of two or more factors may have a weaker or stronger influence on y than each of the individual factors. The q values for the influences of factors x1 and x2 on y, obtained from the application of the factor detector method (Eq. 2), may be represented as q (x1) and q (x2). Hence, a new spatial unit and subregions may be generated by overlaying the factor strata x1 and x2, written as $x1 \cap x2$, where $\cap$ denotes the interaction between factor strata x1 and x2 as illustrated in Figure 3(d). Thus, the q value of the interaction of $x1 \cap x2$ may be obtained, represented as $q\,(x1 \cap x2)$. Comparing the q value of the interaction of the pair of factors and the q value of each of the two individual factors, five categories of the interaction factor relationship can be considered which are summarized in Table 2. If $q(x1 \cap x2) > q(x1) + q(x2)$, this is referred to as a nonlinear enhancement of two variables. And if $q(x1 \cap x2) > \mathrm{Max}[q(x1), q(x2)]$, this is referred to as a bilinear enhancement of two variables. The occurrence of nonlinear enhancement and bilinear enhancement are indicated with the q values in Table 2 and in the caption of Figure 7.".

The occurrence of nonlinear enhancement is indicated with the q values in Table 1 (Table 2 in the revised manuscript) and in the caption of Figure 7 in the revised manuscript.

Table 1. Interaction categories of two factors and the interaction relationship

Illustration	Description	Interaction
	$q(x1 \cap x2) < \mathrm{Min}[q(x1),q(x2)]$	Weakened, nonlinear
	$\mathrm{Min}[q(x1),q(x2)] < q(x1 \cap x2) < \mathrm{Max}[q(x1),q(x2)]$	Weakened, unique
	$q(x1 \cap x2) > \mathrm{Max}[q(x1),q(x2)]$	Enhanced, bilinear
	$q(x1 \cap x2) = q(x1)+q(x2)$	Independent
	$q(x1 \cap x2) > q(x1)+q(x2)$	Enhanced, nonlinear

2. The AOD-CER relationship is difficult to interpret as the Twomey effect, especially where LWP is not controlled for (McComiskey and Feingold, 2012). Several previous studies have also investigated the potential controls on the AOD-CER relationship (e.g. Tan et al, 2017; Yuan et al, 2008; Myhre et al, 2007; Tang et al, 2014; Andersen and Cermak, 2015). There should be a clearer distinction around what is added by this work (which can be the GDM).

Answer: We have taken notice of the references provided and added them to the revised manuscript where appropriate. McComiskey and Feingold (2012) discuss effects of unconstrained LWP, which in the revised manuscript is mentioned in Section 4.2 (lines 442-444): "Note however, that no selection was made for LWP and the condition of constant LWP was not fulfilled. This will be further discussed in Section 4.3." In section 4.3 and Discussions (lines 757-777) we discuss the effect of LWP on S in detail. McComiskey and Feingold (2012) also discuss the spatial separation of satellite observations of cloud and aerosol is addressed in

Section 2.2 (lines 225-229) and a reference to McComiskey and Feingold (2012) has been added: “Aerosol retrieval is only executed in clear sky conditions whereas cloud properties can only be retrieved in cloudy skies. Hence, it is not possible to obtain co-located aerosol and cloud data from satellite. For satellite-based aci studies it is assumed that, following, e.g., Jia et al. (2022), aerosol properties are homogeneous enough to be representative for those in adjacent cloud areas. Consequences of this assumption were discussed by McComiskey and Feingold (2012).”.

Potential controls on the CER-AOD relationship and confounding meteorological effects are discussed in the Introduction, supported with many references (lines 129-160): “Meteorological conditions are important factors determining …. promote the formation of thicker and higher clouds” and in Discussions (lines 778-798): “The above results were obtained by using traditional statistical methods where relationships were derived from scatterplots of CER versus AOD, stratified in two different AOD regimes and five different LWP regimes, as discussed above. The data were also analyzed by using the GDM to determine which factors influence aci and identify how interactions between different parameters influence the results of the aci analysis, i.e. the sensitivity and resulting adjustments. In particular, the GDM provides information on the extent to which the effect of individual factors is influenced by other factors. As shown in Section 4.6.1, the effect of individual factors may be overestimated when confounding effects of other factors are not accounted for. The interaction detector analysis (Section 4.6.2) shows a more realistic estimate of the effects on aci when different factors are analyzed together. The factor detector analysis (Section 4.6.1) shows that over the ECS, AOD has the largest influence on cloud parameters, as indicated by the large and statistically significant q values. Among the meteorological factors, PVV has more influence on the variations of the cloud parameters than RH and LTS. Over the YRD, AOD has the largest influence on COT, with large and significant q values. Among the meteorological factors, the effect of LTS on CF is greater than that of RH and PVV. However, the q-values may sum up to over 100% when the variables are not independent, i.e. the explanatory power of such variables is too high. The evaluation of the effects of interaction between different factors on aci corrects these clearly unrealistic situations. The analysis in section 4.6.2 shows that the interactive q-statistic values derived in this study are larger than any of the values for single variables, i.e. the explanatory power of a combination of factors is higher than that of individual factors, but less than 100%. However, although the GDM provides evidence of the effects of aerosol and meteorological factors and their interactions on cloud properties and quantify the relative contributions to aci, it cannot quantify the absolute contributions with confidence.”.

We have added the references provided by Referee#1 with a brief summary of the findings reported in these references. In Section 6 Conclusions, we have added (lines 807-809): “These results may be influenced by confounding effects of meteorological parameters. The study further shows that over the ECS the CER is larger for higher LTS and RH but lower for higher PVV. Over the YRD, there is no significant influence of LTS on the relationship between CER and AOD.

However, the main comment of Referee #1 is about the question “what is added by this work” and the answer is indeed that it is the GDM as was already mentioned in the Introduction at lines 164-172 in the revised manuscript: “In the current study the geographical detector method (GDM) is applied as a complementary tool to quantify the relative importance of the effects of nine parameters on S. The GDM is explained in detail in Section 3.2. In brief, a set of statistical methods is used to detect the spatial variability of aerosol and cloud properties, which are spatially differentiated, and evaluate the occurrence of correlations in their behaviour and the driving forces behind these correlations (Wang and Hu, 2012; Wang et al., 2016). The basic idea of the GDM is that the spatial distributions of two variables tend to be similar if these two variables are connected (Zhang and Zhao, 2018). The method is used in this study to analyse the relative importance of different factors, and interactions between them, influencing aci."

Furthermore, we have changed the first sentence of Section 3.2 (lines 312-313) to "The geographical detector method (GDM) is introduced to analyze which factors influence the aci and identify possible correlations between different factors" and the text below Figure 2 has been changed (see also response to comment 1).

And in the conclusions we have added the text cited above (lines 810-819): "The GDM has been applied to determine which factors influence S and cloud parameters and the interaction detector analysis has been used to determine the combined effect of different parameters on cloud parameters. The results from the GDM interaction detector analysis clearly show the enhancement of the interaction q-values over the q-values for the individual factors. In other words, the explanatory power of the combined effects of aerosol and a meteorological parameter is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters. We conclude that aerosol and meteorological conditions significantly influence cloud parameters and that combined effects of different factors are often more important than the effect of each individual factor. The relative importance of each factor differs significantly over the ECS and YRD."

3. The majority of more recent studies have used Nd for calculation of the ACI, rather than CER (Quaas et al, 2008; Gryspeerdt et al, 2017; McCoy et al, 2018; Hasekamp et al, 2019). There are also useful studies that investigate the susceptibility (AOD-Nd relationship) and the impacts on this value (Jia et al, 2022). This avoids the LWP-CER issue prevalent in previous work and presents a cleaner separation of the Twomey and adjustments. It could be worth including a section on why CER is used and might be something to consider for future work in this area.

Answer: We address the use of CER rather than N_d with the following text, added to the Introduction (lines 177-186): "It is noted that RF_{aci} is formulated in terms of N_d, whereas studies on the Twomey effects often use CER instead of N_d. CER is readily available as a satellite retrieval product, although in particular over land the reliability is questioned (Grandey and Stier, 2010), whereas N_d is derived from CER and the cloud optical thickness (COT) (e.g., Grandey and Stier, 2010; Arola et al., 2022). This implies that N_d is subject to the same retrieval errors as CER, including a possible relation between CER and LWP. The comparison of global maps of the sensitivities of CER and N_d to AOD by Grandey and Stier (2010) exhibits very similar patterns. In this study, the CER sensitivity to AOD is stratified by LWP, which however poses problems in the evaluation of RF_{aci}. However, the current study focuses on understanding effects of different parameters on CER sensitivity to aerosol rather than the application to determine RF_{aci}.".

4. Previous studies have shown that it is difficult to interpret correlations over large regions as an aerosol effect due to the impact of meteorological confounders (Grandey and Stier, 2010). Correlations between AOD and cloud properties are also fraught with potential confounding effects (Quaas et al, 2010; Boucher and Quaas, 2012; Gryspeerdt et al, 2014). Does the GDM method address these issues? If so how? If not, this study should be much clearer about the claims of causality it puts forward.

Answer: Thank you for this comment. We have added the references provided in the introduction with a brief summary of the findings of each of them. This comment addresses two issues: large regions and confounding effects. As regards large regions: Grandey and Stier (2010) recommend 4°x4° as the largest size and "if data exist at higher gridded resolution the possibility of analyzing data at this higher resolution should be seriously considered." In this study we have followed this recommendation: we have considered two large study regions (YRD and ECS) for the initial evaluation of CER-AOD relationships. Based on these results, we have selected data ranges for which a clear ACI relationship occurs and for these conditions we have refined this study to smaller scales using 1°x1° grid cells and the results are presented in Figure 6 and discussed in Sections 4.2 and 4.3 in the revised manuscript.

We have also considered the size of the study area, or grid size, when using the GDM. In the GDM, the y data are recorded in a raster grid, over a total study area of 9°x9°, as illustrated in Figure 1 (Figure 2 in the manuscript). The data in the raster grid is transformed into dot files, each dot containing a value for y and for one of the influencing parameters x. The dependent (y) and influencing (x) parameters are separated into 2 layers with the same grid. As the resolution of MYD 08 data used in this study is 1°x1°, the data transformed into dot files is based on raster grid 1°x1°. Here, 15-year averaged distributions of clouds (y, 5 layers) and aerosols/meteorological conditions (x, 4 layers) are used as input in the GDM. Tables 2 and 3 show the q values for factors which may influence cloud parameters over the ECS and YRD in different regions sizes, evaluated for data collected in the period from 2008-2022. The data in Tables 2 and 3 show that for regions smaller than 9°x9°, the GDM result is not significant and that the results become more significant when the area is getting larger (for example 9°x9°, 10°x10°, 11°x11°, 12°x12°). In future research, higher resolution data can be used for GDM by controlling the size of the study area to be less than 4°x4°.

The problem of large regions and the effect of possible meteorological confounders was also addressed by Arola et al. (2022) and in response to a comment by Referee#2 we have added the following text to Section 5 (Discussion, lines 757-777): "Arola et al. (2022) addressed the susceptibility of N_d to changes in aerosol and the adjustment of LWP (using satellite observations), and confounding factors, in particular co-variability of N_d and LWP induced by meteorological effects. They show how errors in the retrieved CER and COT or spatial heterogeneity in cloud fields influence the N_d - LWP relation. However, both N_d and LWP are not retrieved but derived from CER and COT. Using Eq. 1 and Eq. 2 in Arola et al. (2022), the N_d -LWP relationship can be shown to have a highly non-linear dependence on CER and thus it is no surprise that any error in CER strongly affects the relation between N_d and LWP. Their experiments, i.e. using smaller scales (5° x 5°) to reduce spatial meteorological variability, or using snapshots to remove meteorological variability in time, did not lead to a conclusion whether the N_d - LWP variability is due to spatial heterogeneity in the cloud fields or due to retrieval errors. The main message from this part of the study (using satellite data) by Arola et al. (2022) is "the spatial variability of CER introduces a bias which moreover becomes stronger in conditions where the CER values are lower on average". Experiments with simulated measurements show that "the main cause of the negative LWP vs N_d slopes is the error in CER". Arola et al. emphasize that the spatial cloud variability and retrieval errors in CER and COT are similar sources for negative bias in LWP adjustment and that these sources could not be separately assessed in their simulations. The implication of the findings of Arola et al. (2022) on the adjustment of LWP for the results of the current study on the sensitivity of CER to aerosol (or CCN, using AOD as proxy) is that the assumption of constant LWP may be violated. This would affect the results presented in Section 4.3 where LWP was stratified and S was found to vary with LWP. In view of the LWP adjustment to changes in aerosol, the variation of CER sensitivity with LWP may be somewhat different from that reported in section 4.3."

The results from the GDM interaction detector analysis in Section 4.6.2 clearly show the enhancement of the interaction q-values over the q-values for the individual factors. In other words, the explanatory power of the combined effects of a meteorological parameter and aerosol is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters.

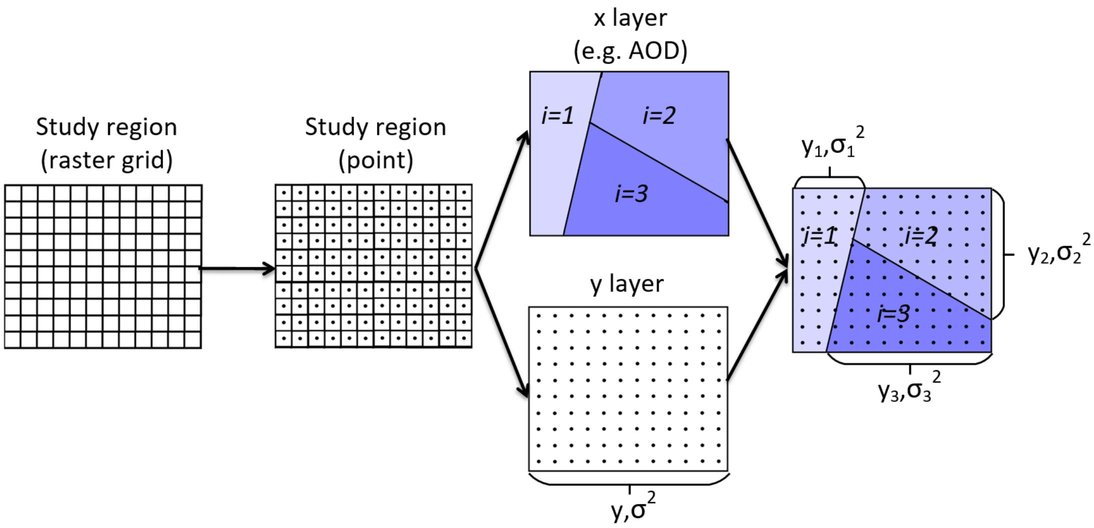

Figure 1. The principle of the geographical detector method. See text for explanation.

Table 2. q values for factors which may influence cloud parameters over the ECS in areas with different sizes, evaluated for data collected in the period from 2008-2022.

Region	Cloud	AOD	RH	LTS	PVV
4°x4°	CER	0.67	0.43	0.14	0.59
	COT	0.27	0.51	0.28	0.44
	LWP	0.72	0.25	0.09	0.53
	CF	0.68	0.29	0.11	0.28
	CTP	0.31	0.50	0.29	0.57
5°x5°	CER	0.79	0.42	0.40	0.72
	COT	0.54	0.41	0.25	0.48
	LWP	0.57	0.37	0.34	0.52
	CF	0.46	0.48	0.16	0.53
	CTP	0.49	0.45	0.25	0.31
6°x6°	CER	0.85^{**}	0.47	0.49	0.54
	COT	0.62	0.62	0.37	0.56
	LWP	0.64	0.34	0.41	0.31
	CF	0.30	0.23	0.16	0.40
	CTP	0.46	0.47	0.22	0.45
7°x7°	CER	0.73^{***}	0.36	0.39	0.63^{***}
	COT	0.60	0.37	0.37	0.53^{**}
	LWP	0.58	0.19	0.31	0.47
	CF	0.34	0.13	0.05	0.43^{**}

	CTP	0.37	0.47	0.21	0.47
	CER	0.86^{***}	0.48	0.57^{***}	0.63^{**}
	COT	0.72^{***}	0.50	0.54	0.65^{***}
8°x8°	LWP	0.71	0.41	0.48	0.51
	CF	0.39	0.27	0.14	0.42^{**}
	CTP	0.48	0.56	0.56	0.56
	CER	0.81^{***}	0.33^{***}	0.44^{***}	0.70^{***}
	COT	0.69^{***}	0.40	0.38	0.67^{***}
9°x9°	LWP	0.68^{***}	0.23	0.43^{***}	0.49^{***}
	CF	0.46^{***}	0.20	0.09	0.47^{***}
	CTP	0.47	0.53	0.18	0.58
	CER	0.86^{***}	0.46^{***}	0.54^{***}	0.62^{***}
	COT	0.71^{***}	0.55^{**}	0.52^{***}	0.67^{***}
10°x10°	LWP	0.72^{***}	0.29	0.39^{***}	0.47^{***}
	CF	0.49^{***}	0.19	0.09	0.44^{***}
	CTP	0.53	0.58	0.29	0.66
	CER	0.87^{***}	0.45^{***}	0.39^{***}	0.54^{***}
	COT	0.71^{***}	0.53^{***}	0.45^{***}	0.60^{***}
11°x11°	LWP	0.73^{***}	0.73	0.26	0.44^{***}
	CF	0.48^{***}	0.13	0.04	0.29^{***}
	CTP	0.62	0.52	0.26	0.57
	CER	0.84^{***}	0.42^{***}	0.31	0.55^{***}
	COT	0.66^{***}	0.46^{***}	0.37	0.52^{***}
12°x12°	LWP	0.64^{***}	0.30	0.05	0.41^{***}
	CF	0.42^{***}	0.13	0.11	0.24^{***}
	CTP	0.53	0.49	0.27	0.54

Note: ***indicates that the q value is significant at the 0.01 level ($p < 0.01$), **indicates that the q value is significant at the 0.05 level ($p < 0.05$).

Table 3. q values for factors which may influence cloud parameters over the YRD in areas with different sizes, evaluated for data collected in the period from 2008-2022.

Region	Cloud	AOD	RH	LTS	PVV
	CER	0.47	0.64	0.04	0.27
	COT	0.75	0.62	0.37	0.55
4°x4°	LWP	0.60	0.54	0.49	0.60
	CF	0.42	0.62	0.06	0.62
	CTP	0.85	0.59	0.53	0.77
	CER	0.28	0.53	0.24	0.17
	COT	0.79	0.43	0.43	0.53
5°x5°	LWP	0.69	0.49	0.44	0.38
	CF	0.46	0.42	0.33	0.51
	CTP	0.86	0.69	0.57	0.60
	CER	0.17	0.14	0.18	0.11
	COT	0.75	0.30	0.27	0.41
6°x6°	LWP	0.71	0.32	0.31	0.26
	CF	0.30	0.34	0.12	0.39
	CTP	0.81	0.53	0.40	0.54

7°x7°	CER	0.18	0.21	0.06	0.19
	COT	0.75	0.47	0.21	0.53
	LWP	0.43	0.44	0.52	0.33
	CF	0.36	0.26	0.13	0.23
	CTP	0.73	0.75	0.46	0.65
8°x8°	CER	0.31	0.24	0.34	0.17
	COT	0.66***	0.45	0.24	0.31
	LWP	0.21	0.43	0.60	0.38
	CF	0.28**	0.07	0.68***	0.05
	CTP	0.60	0.75	0.45	0.56
9°x9°	CER	0.31	0.25	0.13	0.18
	COT	0.61***	0.45***	0.12	0.29
	LWP	0.16	0.32	0.55***	0.18
	CF	0.30***	0.02	0.50***	0.07
	CTP	0.50	0.74***	0.32	0.56
10°x10°	CER	0.41	0.28	0.20	0.27
	COT	0.63***	0.50**	0.08	0.37
	LWP	0.21	0.36	0.51***	0.22
	CF	0.38***	0.06	0.48***	0.17
	CTP	0.50	0.78	0.31	0.60
11°x11°	CER	0.35	0.28	0.17	0.22
	COT	0.69***	0.40***	0.06	0.46
	LWP	0.35**	0.28	0.40	0.24
	CF	0.39***	0.06	0.47***	0.15
	CTP	0.48	0.72***	0.24	0.49
12°x12°	CER	0.32	0.19	0.19	0.16
	COT	0.50***	0.28***	0.07	0.47
	LWP	0.18**	0.25***	0.36***	0.26
	CF	0.37***	0.06	0.45***	0.12
	CTP	0.32	0.65***	0.25	0.35

Note: ***indicates that the q value is significant at the 0.01 level ($p < 0.01$), **indicates that the q value is significant at the 0.05 level ($p < 0.05$).

5. Another important factor is the calculation of LWP that is used for binning in the ACI calculations. As the LWP depends on the CER, does this not lead to an implicit filtering by CER, which would affect the calculation of ACI?

Answer: Stratification of the data for LWP was applied by, e.g., Saponaro et al. (2017) and Ma et al. (2018) in an attempt to satisfy the conditions for the Twomey effect. Indeed, Ma et al. (2018) show that the variation of the CER vs AI (both stratified according to LWP) relation changes with changes in LWP. The data in Section 4.3 also show the variation of S with the LWP interval and likely this is a more continuous variation if smaller LWP intervals (quasi-constant LWP, Ma et al., 2018) would be used. So indeed the assumption of constant LWP is not truly satisfied and this indeed affects the calculation of ACI as can be deduced from the data in Table 4 below (Table 3 in the revised manuscript). We further note that Arola et al. (2022) and others show a clear LWP - N_d relationship, in agreement with other studies. And LWP and N_d are both calculated from CER and COT, so a relationship is expected. We have addressed the findings by Arola et al. (2022) and this text was copied in our response to comment 4. We have added the following text in Section 4.3 (lines 528-534): “The variation of S with changes in LWP indicates that the condition of constant LWP is not truly satisfied: if the data would be stratified according to smaller LWP intervals (quasi-constant LWP, Ma et al., 2018), S would likely vary more smoothly with LWP. As mentioned in the Introduction, LWP is not directly retrieved but calculated form CER and COT and thus also the calculation of S is to some extend affected by LWP. We further note the results by Ma et al. (2018), i.e. the slope of CER versus AI (comparable to S in this paper) varies little with LWP, with positive values over land and negative values over ocean and thus behaves similar to the data in Table 4 (Table 3 in the revised manuscript) for YRD and ECS.”.

Table 4. Estimates of S, computed using Eq. (1), and correlation coefficients R between CER and AOD, stratified by LWP, over the ECS for 0.1<AOD<0.3 and over the YRD for AOD>0.3. Statistically significant data points are indicated with * (p value < 0.01).

	ECS (0.1<AOD<0.3)		YRD (AOD>0.3)	
LWP (g m^{-2})	S	R	S	R
0-40	0.10	0.94*	0.08	0.63*
40-80	-0.19	-0.98*	0.10	0.81*
80-120	-0.38	-0.99*	0.06	0.57*
120-160	-0.41	-0.99*	-0.03	-0.11
160-200	-0.46	-0.98*	-0.14	-0.42*

6. Is there a reason for using AOD, rather than a product such as the aerosol index (Nakajima et al, 2001), which has a stronger link to the CCN concentration?

Answer: AI=AOD*AE, but AE retrieval over land from MODIS is problematic (Ma et al., 2018 refers to Sayer et al., 2013) and therefore is no longer provided as a MODIS product in C6! We cite Ma et al.: “using AOD instead of AI does not influence the conclusions. (next to their Table 1)”

Another argument may come from Gryspeerdt et al. (2023): “The larger relative error in the aerosol retrieval under clean conditions reduces the correlation between the CCN and the retrieved aerosol due to regression dilution (Pitkänen et al., 2016). This reduces the magnitude of β under clean conditions, as observed in Fig. 1a and b. This issue is particularly severe for AI, which is calculated using the ratio of aerosol optical depths at two wavelengths, resulting in a relative error which tends to infinity under clean conditions” (β = d ln Nd / d Ln A, where A is the aerosol proxy AI or AOD). The problem occurs under clean aerosol conditions because the contribution of the surface to the TOA result in larger uncertainty in the retrieved AOD.

We have added the following text in the Introduction (lines 106-122): “In studies on S utilizing satellite data, which is the subject of the current study, the aerosol optical depth (AOD) is often used as a proxy for the aerosol concentration, which is justified by the correlation of AOD and CCN published by Andreae (2009). However, AOD is determined by all aerosol particles in the atmospheric column, including particles that do not act as CCN, depends on the relative humidity (RH) throughout the atmospheric column, does not provide information on chemical composition and may be influenced by aerosol in disconnected layers. The use of the Aerosol Index (AI), the product of AOD and the Ångström Exponent (AE; describing the spectral variation of AOD), is suggested as a better indicator of CCN because AE includes information on aerosol size (e.g., Nakajima et al., 2001). However, the AE is determined from AOD retrieved at two or more wavelengths and the evaluation of the results versus ground-based reference data shows the large uncertainty in AE. Therefore, in recent MODIS product Collections, AE is not provided over land (e.g., Levy et al., 2013; Kourtidis et al., 2015). AE is also not well-defined for low AOD for which uncertainty is largest (Bellouin et al., 2020; Gryspeerdt et al., 2023). The issues associated with using AOD or AI as proxy for CCN were discussed by, among others, Rosenfeld et al. (2014) who do not recommend the use of AI while also concluding that no better proxy is available. Therefore, in this study, AOD is used as a proxy for CCN to study S. It is noted that in other studies, e.g., Jia et al., 2022, both AOD and AI have been used and the results show similar behaviour.".

Specific points

1. The abstract mostly list results, rather than providing an overview of the paper and the conclusions. Is there an overall picture or aim of the study that could help to structure this?

Answer: We have revised the abstract substantially and added to following sentence upfront, to provide the overall picture "The sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters has in most previous aerosol-cloud interaction (aci) studies been addressed using traditional statistical methods. In the current study, relationships between cloud droplet effective radius (CER) and aerosol optical depth (AOD, used as a proxy for cloud condensation nuclei, CCN), i.e. the sensitivity (S) of CER to AOD, is investigated with different constraints of AOD and cloud liquid water path (LWP). In addition to traditional statistical methods, the geographical detector method (GDM) has been applied to quantify the relative importance of the effects of aerosol and meteorological parameters, and their interaction, on S." Note that many other changes were made to the abstract in "track changes".

2. L39 - Is this opposite effect just because the sign of the pressure vertical velocity is defined differently? I am not sure what opposite means in this context.

Answer: We can see that the CER decreases with increasing AOD over the ECS, which is consistent with the Twomey effect. The meteorological parameters do no change the trend of CER variation to the AOD. However, the CER is larger for higher LTS and RH but lower for higher PVV. We also reorganized the text with "The study further shows that over the ECS the CER is larger for higher LTS and RH but lower for higher PVV." (see lines 32-33) in the revised manuscript.

3. L68 - The terms first and second indirect effect are less commonly used in more recent studies. I would suggest referring to adjustments instead (see IPCC AR5), as this more closely links in with the radiative forcing/effective radiative forcing distinction and aligns more closely with other recent work.

Answer: Thank you for this valuable comment. We have changed the terminology throughout the revised manuscript and used several key references to guide us, such as IPCC AR5, Gryspeerdt et al. (2023), Bellouin et al. (2020) and several others.

4. L81 - I would have said that satellites typically have a fairly poor temporal resolution (unless the authors are referring to geostationary satellites?)

Answer: In this paper we only use MODIS data. We have removed "and high spatial and temporal resolution"

5. L93 - Is there any reason for choosing these studies? They seem to be rather disjointed, with some looking at the Twomey effect directly and some considering adjustments. Some notable studies looking at the impact of meteorological parameters on potential adjustments (e.g. Koren et al, 2010) and the particularly Twomey effect (Jones et al, 2009; Jia et al. 2022) are left out.

Answer: We have reorganized the text and added the notable references looking at the impact of meteorological parameters on potential adjustments (e.g. Koren et al, 2010) and the particularly Twomey effect (Jones et al, 2009; Jia et al. 2022）. See the text on pages 5-6 (lines 129-164) in the revised manuscript.

6. L96 - PVV is redefined here

Answer: Consistent notation has been used through the revised manuscript.

7. L116 - There needs to be some discussion of how the GDM is affected by the results of Grandey and Stier (2010), who suggest that spatial correlations are unreliable. It may be that the results of GS10 are not applicable here, as the GDM method is capable of accounting for the co-variations that drive the results in GS10. If so, it would be good to have some evidence of this, as it would provide more significance to the results presented in this work.

Answer: Spatially-varying aerosol and cloud properties may contribute towards observed relationships between aerosol and cloud properties. This may affect the results of many of the aforementioned studies which analyze data on a relatively large regional scale. Aerosol type, cloud regime and synoptic regime may vary over such large spatial scales. If data are analyzed for the region as a whole, false correlations may be introduced. Grandy and Stier (2010) suggested that for region sizes larger than 4°x4°, spurious spatial variations in retrieved cloud and aerosol properties can introduce widespread significant errors to calculation S. However, we can observe that at the regional scales of 8°x8° and 15°x15°, although significant errors are introduced, the spatial distribution patterns of S (the sensitivities of CER and N_d to AOD) look very similar, as shown in Figure 2 of Grandy and Stier (2010).

GDM is a spatial statistical analysis method aimed at studying the degree of influence and spatial patterns of different factors on the changes in geographic phenomena. In the analysis, we can simultaneously consider the interactions and impacts among multiple factors, thus revealing the relationships of synergistic changes. Therefore, the geographic detector method can encompass the analysis of co-variations.

We have added the following text in the Sect 4.6.1 (lines 647 to 666): "Tables 5 and 6 list q values for individual factors, together with p showing the absence of statistical significance in many cases, especially over the YRD, and often the explanatory power is not high when the significance is low. These data show that cloud parameters are dominated by aerosol effects over the ECS but meteorological influences on cloud parameters predominate over the YRD, as was also concluded from the analysis from "traditional" statistical methods presented in Section 4.5 and these conclusions are consistent with the results published by Andersen and Cermak (2015). Among the meteorological parameters, we also find that PVV (with highest q in the three meteorological parameters) predominantely influences cloud parameters over the ECS. Jones et al. (2009) and Jia et al. (2022) reported that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. In addition, we find that CTP is mainly affected by RH ($q = 0.74^{***}$) and PVV ($q = 0.56$) over the YRD, as suggested by Koren et al. (2010). Koren et al. reported that observed cloud top height correlates best with model pressure updraft velocity and relative humidity. To some extent, LTS influences CER ($q = 0.44^{***}$) and LWP ($q = 0.43^{***}$) over the ECS, while, in contrast, over the YRD LTS predominately influences CF ($q = 0.50^{***}$) and LWP ($q = 0.55^{***}$). Matsui et al. (2004) and Tan et al. (2017) reported that aerosol impact on CER is stronger in more dynamic environments that feature a lower LTS and argue that very high LTS environments dynamically suppress cloud droplet growth and reduce aci intensity. While strong correlations between AOD and cloud parameters have been previously observed, they are likely due to the swelling of aerosol particles in humid airmasses (Quaas et al, 2010), rather than an aerosol influence, which is in agreement with findings by, e.g., Myhre et al. (2007), Twohy et al. (2009) and Quaas et al. (2010)."

This study provides a general description of the sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters over the YRD and the ECS. Correlations between AOD and cloud parameters are found over the target regions, which can be attributed in part to the influence of general circulation. In general, there are many relations between the various parameters, both related to cloud microphysics and meteorology. Thus, establishing cause and effect relationships between parameters is difficult and must be made with care. It is not possible to completely separate meteorological influences from aerosol influences on clouds. This work can therefore only provide further evidence of the aerosol and meteorological effect on clouds and quantify the relative contributions and combined effects on clouds, but cannot quantify the absolute contributions with confidence.

In the current study, based on a regional scale of 9°x9°, the GDM method is used to explore the relative importance of various factors on cloud parameters and identify possible correlations between different factors. In the future, aerosol cloud interactions can be studied on smaller regional scale (<4°x4°) using higher resolution source data.

8. L154 - Why only 2008 to 2022? The MODIS record runs back to 2002/3

Answer: There is no particular reason for the selection of 2008 as the starting year. Most other studies use a shorter period of time. Based on your comment and that of other referees, we have thought about shorter periods and realized that, in principle, periods were included when the AOD was at its maximum (2008-2014) and when the AOD was decreasing in response to implementation of emission reduction policy. We therefore split the data sets for these 2 periods and plotted CER vs AOD, see Figures 2 and 3 below. We noticed that over the ECS there was not a significant difference between the CER/AOD relations during these two periods. However, over the YRD, for the high AOD period, CER clearly decreased with increasing AOD for 0.1<AOD<0.3 and for larger AOD the CER increased with R=0.87. For the second period, however, there was no clear correlation between CER and AOD for both AOD intervals. The data also show that over the YRD the CER for AOD>0.3 increased to larger values during the first period than during the second period. We did not look for explanations of this difference, possibly the aerosol properties changed in response to emission reduction, or confounding meteorological factors played a role.

We also looked for shorter periods, considering each year between 2008 and 2022. The results show similar behavior for each year over both study areas with interannual variations between the fits, and thus the value of S. However, the statistical significance is low (large p) due to the small number of samples.

These findings were briefly summarized in the Discussion (lines 733-746): “These results were obtained using data from a period of 15 years. During this period, the aerosol properties changed in response to expanding economy, resulting in the increase of the AOD until 2007, and the implementation of emission reduction policy resulting in the decrease of the AOD from 2014 which flattened from about 2018 (de Leeuw et al., 2021; 2022; 2023). To account for these changes, the sensitivity S was determined for the periods 2008-2014 and 2014-2022, without stratification for LWP (see Figures S1 and S2 in the Supplementary). The results for the ECS show no significant difference between the CER-AOD relations during these two periods. Over the YRD, however, the data for 2008-2014 show a clear decrease of CER with increasing AOD for 0.1<AOD<0.3 and for larger AOD the CER increased, with a statistical significant correlation (R=0.87) and S=0.10 as compared to S=0.08 for the whole period. In contrast, the data for 2014-2022 show no clear correlation between CER and AOD for both AOD intervals over the YRD. A similar exercise for shorter periods, i.e. for each year between 2008 and 2022, show similar behavior as for the whole period 2008-2022, over both study areas, with interannual variations of the value of S. However, the statistical significance is low (large p) due to the small number of data samples in each year.”

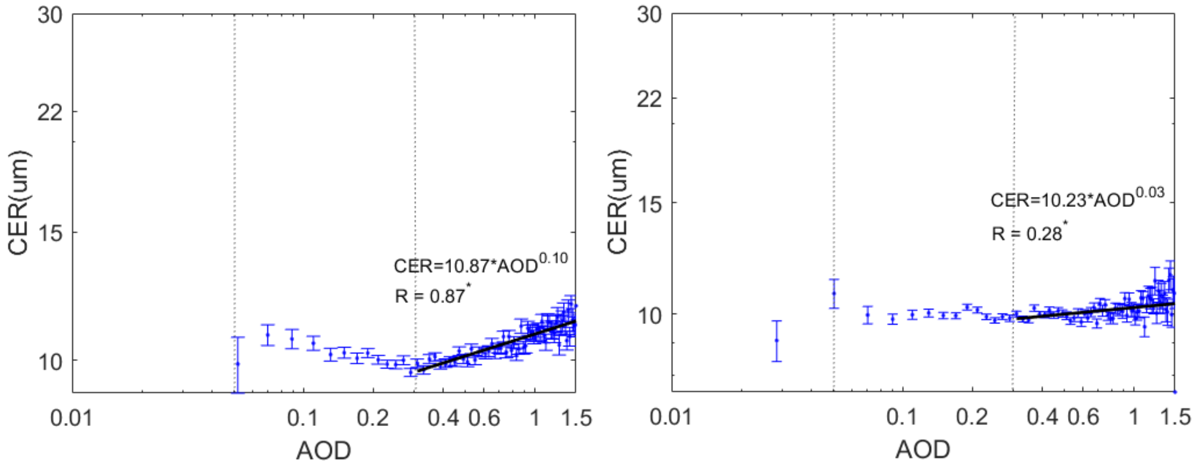

Figure 2. CER vs AOD over the YRD for the periods 2008-2014 (left) and 2015-2022 (right).

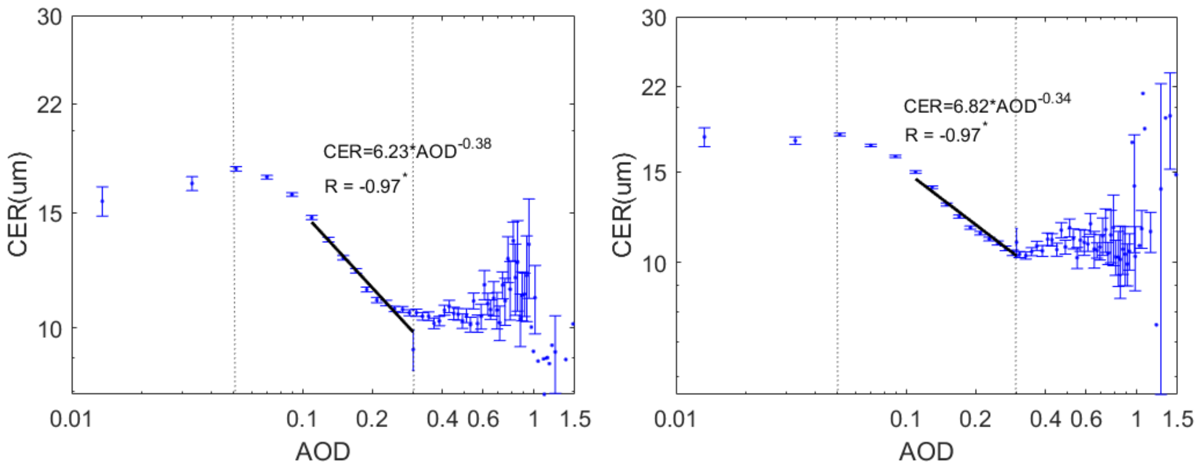

Figure 3. CER vs AOD over the ECS for the periods 2008-2014 (left) and 2015-2022 (right).

9. L159 - The aerosol and cloud retrievals are necessarily conducted in different regions of the 1x1 degree gridbox (aerosol retrievals are only conducted in clear sky), meaning that they are not coincident. This may not affect the results if the regions are non-precipitating. Jia et al (2022) showed that wet scavenging can have a considerable impact on the susceptibility.

Answer: The use of non-collocated aerosol and cloud data is addressed in Section, 2.2. (lines 225-229): "Aerosol retrieval is only executed in clear sky conditions whereas cloud properties can only be retrieved in cloudy skies. Hence, it is not possible to obtain co-located aerosol and cloud data from satellite. For satellite-based aci studies it is assumed that, following, e.g., Jia et al. (2022), aerosol properties are homogeneous enough to be representative for those in adjacent cloud areas. Consequences of this assumption were discussed by McComiskey and Feingold (2012)."

We have filtered the data (exclude precipitating clouds) following the method used in Ma et al. (GRL2018) and Saponaro et al. (2017) (see lines 265-267 in the revised manuscript): "To ensure that the data used only included single layer liquid clouds and nonprecipitating cases, the filtering criteria described by Saponaro et al. (2017) were applied.". This issue is shown throughout the revised manuscript (all the figures were changed/modified in this respect).

10. L153 - I would suggest referencing Platnick et al (2016), given the authors are using MODIS collection 6.1.

Answer: We have added the following text to Section 2.2. (lines 236-245): "The MODIS Collection 6.1 AOD product over China has been validated by, e.g., Che et al. (2019) and globally over land and ocean by Wei et al. (2019). MODIS C6.1 cloud products were evaluated by Platnick et al. (2017). The validation of CER and LWP, the primary cloud products used in this paper, was described by Painemal and Zuidema (2011), who compared MODIS C5 with in situ data (aircraft), and likewise the MODIS C6.1 CER product was evaluated by Fu et al. (2022) by comparison with airborne measurements. Fu et al. (2022) concluded that their "validation, along with in situ validation of MODIS CER from other regions (e.g., Painemal and Zuidema, 2011; Ahn et al., 2018), provides additional confidence in the global distribution of bias-adjusted MODIS CER reported in Fu et al. (2019)." It is noted that COT and CER are retrieved whereas LWP is secondarily derived (e.g., Painemal and Zuidema, 2011)."

11. L169 - Brendan et al (2005) suggests that cloud contamination becomes an issue when the AOD is larger than 0.6. Why is a larger threshold used here?

Answer: The conclusion of Brendan et al. (2005) applies to the MOD06 Collection 04 cloud product and these authors conclude with "The cloud masking technique in the recently updated Collection 05 cloud retrieval algorithm has been improved, and the Collection 05 cloud products available in the near future will largely eliminate the aerosol contamination effect". Christenson et al. (ACP 2017) used MOD06 C6 data (1km x1km) and reported that "large aerosol optical depths remain in the MODIS-observed pixels near cloud edges, due primarily to 3-D effects (Varnái and Marshak, 2009) and the swelling of aerosols by higher relative humidity." And "Varnái and Marshak (2009) also noted that beyond 15 km contamination effects were minimized in MODIS data (1km x1km)." Therefore Christensen et al. only used data pairs beyond the 15 km length scale in their aci study.

In our study we use MODIS L3 collection 6.1 with a spatial resolution of 1°x1°. Comparisons with surface-based sun photometer data revealed that Collection 6 should improve upon

Collection 5, and overall, 69.4% of MODIS Collection 6 AOD fell within an expected uncertainty of ± (0.05 + 15%) (Levy et al., 2013; Tan et al., 2017). In this study, to eliminate 1° by 1° scenes in which the aerosol distribution is heterogeneous, retrievals with a standard deviation higher than the mean values are discarded (Saponaro et al., 2017; Jia et al., 2022). In addition, many previous researches do not set a threshold of AOD when using MODIS L3 C6 data (Grandey and Stier, 2010; Tang et al., 2014; Saponaro et al., 2017; Tan et al., 2017; Ma et al., 2018; Jia et al., 2022). Based on these findings, we used the larger threshold of 1.5.

These explanations have been summarized in the text added to section 2.2 (lines 252-260): "The choice of this threshold, rather than 0.6 used by Brendan et al. (2006), who used MOD06 Collection 04 products, is based on reports by Christenson et al. (2017) and (Varnái and Marshak, 2009). Christenson et al. (2017) used MOD06 C6 data (1km x1km) and reported that "large aerosol optical depths remain in the MODIS-observed pixels near cloud edges, due primarily to 3-D effects (Varnái and Marshak, 2009) and the swelling of aerosols by higher relative humidity." Varnái and Marshak (2009) noted that beyond 15 km contamination effects were minimized in MODIS data (1km x1km). Furthermore, we discarded scenes (1° by 1°) in which the aerosol distribution is heterogeneous, i.e. with a standard deviation higher than the mean value (Saponaro et al., 2017; Jia et al., 2022)."

12. L172 - Why is 200gm^-2 used as a threshold for the LWP?

Answer: In the text we added (line 261): "LWP larger than 200 g m^{-2} is excluded to avoid deep convective clouds (Wang et al., 2014)".

13. L184 - I would suggest putting the URL links in the references or acknowledgements

Answer: Although it is nowadays quite common to provide url + last accessed data as a reference in the text, we have followed this suggestion.

14. L187 - ERA5 and ERA Interim both seem to be mentioned at different points in this work. I suggest using only one (preferably ERA5)

Answer: Indeed we used ERA-5 and have corrected this throughout the text.

15. L190 - I am not sure this is the definition of the first indirect effect as all of these properties also vary with cloud adjustments.

Answer: In response to your comments and those from other reviewers, we have changed the terminology to the terminology recommended in IPCC AR5 (see also our response to your specific point 3). As a result, we have change the title of Section 3.1 to "Sensitivity of cloud parameters to changes in aerosol concentrations" and the first sentence now reads "Changes in aerosol loading lead to an adjustment of cloud optical or microphysical parameters (COT, CER, etc.).", together with many other changes throughout the revised manuscript.

16. L192 - Ice nuclei are usually referred to as "ice nucleating particles" (INP) - Vali et al (2015)

Answer: Thank you for this suggestion: this has been changed here and elsewhere in the revised manuscript.

17. L204 - Do the authors mean CCN here (as in Andreae, 2009)

Answer: In the original formulation by Feingold et al (2001), α is the AOD. This relation was derived assuming that the cloud droplet number concentration N_d varies with the aerosol number as N_a^{a1}(their eq. 5), with a1=0.7. Following Andreae (2009) there is a power law relation between AOD and CCN. We changed the sentence below eq. 1 (see lines 300-301 in the revised manuscript) to "Where r_e represents the CER and α represents the AOD. Following Andreae (2009), AOD and CCN are correlated and AOD varies with CCN following a power law relationship.", while also changes were made to the rest of this paragraph.

18. L216 - An explicit list of these parameters, perhaps in the diagram, could be useful for others trying to replicate this study.

Answer: Thank you for this comment. We have added this information in Table 5 (Table 1 in the revised manuscript).

Table 5. Parameters used in the present study, together with the sources, time periods and spatial resolutions.

Source	Time period	Resolution	Parameters
MYD08	Jan 2008-Dec 2022	Daily, 1°x1°	AOD at 550 nm
			COT at 2.1 um
			CER at 3.7 um and 2.1 um
			Cloud-top temperature
			Cloud-top pressure
			LWP at 2.1 um
			Cloud Fraction
			Solar zenith angle
			Sensor zenith angle
			Cloud multi-layer flag
			Cloud phase flag
ERA5	Jan 2008-Dec 2022	hourly, 0.25°x0.25°	Temperatures at 700 and 1000 hPa
			Relative humidity at 750 hPa
			Vertical velocity at 750 hPa

19. L221 - I have not used the Jenks method before, but from what I understand, you have to specify the number of regions/regimes? How is this done and does the number of regions chosen affect the results?

Answer: The geographic detector model requires the input independent variable to be a type variable. The Jenks natural breaks classification method is one of many discretization methods and is commonly used in literature. The Jenks natural breaks classification method (Brewer and Pickle, 2002), aiming to minimize the variance within the group and maximize the variance between groups, was applied to categorize the whole region into n subregions. For example, AOD needs to be classified into 5 levels using the Jenks natural breaks classification method, and the AOD source data needs to be reclassified into 1-5 natural numbers from small to large, and then counted into the grid. Therefore, the input of the independent variable AOD is a type variable. However, it should be noted that the GDM also has unstable characteristics. On the one hand, it is due to the MAUP (Modified Area Unit Problem) variable area unit problem, which can be understood as the influence of "scale effect". Due to the limitation of data resolution used in this study, the spatial statistical unit is 1°x1°. On the other hand, the methods used for data discretization can also have an impact. This study attempts to determine the optimal number of classifications by examining the impact of different classification numbers (3-8) on the GDM output results (as shown in Tables 6 and 7 below). The data shows that the classification number of regions does not affect the relative importance of cloud factors on the cloud. Here we classify the values of each cloud factor into 5 levels during the period of 2008-2022.

Table 6. q values for factors which may influence cloud parameters over the ECS (9°x9°) in different number of classification levels (3~8) (see text) using Jenks natural breaks classification method, evaluated for data collected in the period from 2008-2022.

cloud parameters	number of classification levels	AOD	RH	LTS	PVV
CER	3	0.80***	0.33**	0.42***	0.69***
	4	0.81***	0.40***	0.43***	0.67***
	5	0.81***	0.33**	0.44***	0.70***
	6	0.85***	0.41	0.52***	0.73***
	7	0.83***	0.37	0.44	0.74***
	8	0.84***	0.40	0.48**	0.70***
COT	3	0.66***	0.43	0.42**	0.64***
	4	0.69***	0.45	0.43	0.66***
	5	0.69***	0.40	0.38	0.67***
	6	0.72***	0.47	0.50	0.72***
	7	0.75***	0.49	0.43	0.71***
	8	0.75***	0.48	0.46	0.68***
LWP	3	0.68***	0.18	0.34***	0.57***
	4	0.67***	0.25	0.37**	0.48***
	5	0.68***	0.23	0.43***	0.49***
	6	0.72***	0.27	0.44	0.55***
	7	0.71***	0.30	0.36	0.59***
	8	0.75***	0.26	0.45	0.58***
CF	3	0.42***	0.19	0.05	0.46***
	4	0.46***	0.18	0.07	0.44***
	5	0.46***	0.20	0.09	0.47***
	6	0.47***	0.22	0.07	0.50***
	7	0.49***	0.19	0.08	0.56***
	8	0.49***	0.22	0.09	0.50***
CTP	3	0.47	0.48	0.24	0.60
	4	0.44	0.56	0.21	0.58
	5	0.47	0.53	0.18	0.58
	6	0.51	0.56	0.36	0.69
	7	0.50	0.57	0.27	0.66

	8	0.51	0.58	0.26	0.65

Note: ***indicates that the q value is significant at the 0.01 level ($p < 0.01$), **indicates that the q value is significant at the 0.05 level ($p < 0.05$).

Table 7. q values for factors which may influence cloud parameters over the YRD (9°x9°) in different number of classification levels (3~8) (see text) using Jenks natural breaks classification method, evaluated for data collected in the period from 2008-2022.

cloud parameters	number of classification levels	AOD	RH	LTS	PVV
CER	3	0.22	0.14	0.01	0.12
	4	0.32	0.19	0.05	0.14
	5	0.31	0.25	0.13	0.18
	6	0.33	0.17	0.17	0.23
	7	0.34	0.25	0.17	0.15
	8	0.38	0.27	0.19	0.23
COT	3	0.52***	0.47**	0.08	0.19
	4	0.53***	0.52***	0.10	0.31
	5	0.61***	0.45	0.12	0.29
	6	0.56**	0.45	0.11	0.28
	7	0.60***	0.49	0.12	0.28
	8	0.59	0.54	0.15	0.32
LWP	3	0.17	0.35	0.52***	0.16
	4	0.17	0.34	0.54***	0.00
	5	0.16	0.32	0.55***	0.18
	6	0.18	0.34	0.55	0.21
	7	0.18	0.38	0.54**	0.18
	8	0.23	0.37	0.55	0.20
CF	3	0.30***	0.01	0.34***	0.04
	4	0.37***	0.02	0.45***	0.03
	5	0.30***	0.02	0.50***	0.07
	6	0.39***	0.03	0.50***	0.09
	7	0.36***	0.05	0.58***	0.06
	8	0.38***	0.04	0.56***	0.10
CTP	3	0.49	0.72**	0.26	0.48
	4	0.46	0.74***	0.35	0.52
	5	0.50	0.74***	0.32	0.56
	6	0.52	0.75	0.32	0.56
	7	0.55	0.79	0.38	0.57
	8	0.50	0.79	0.36	0.56

Note: ***indicates that the q value is significant at the 0.01 level ($p < 0.01$), **indicates that the q value is significant at the 0.05 level ($p < 0.05$).

20. Eq2 - I am not familiar with this method, so might need a bit more explanation. Is sigma here the variance of y within the specified region/regime?

Answer: Sigma σ here is the standard deviation of y within the specified region/regime and σ^2 is the variance of y within the specified region/regime. This is specified in the text on page 13 of the revised manuscript (see lines 334-335): "and σ_i^2 and σ^2 denotes variance of samples in the subregion i and the total variance in the entire study area, respectively.".

21. Eq2 - How does this method compare to a more common correlation measure for non-linear relationships, such as Spearman's Rank?

Answer: Spearman's Rank analysis and GDM are two different statistical methods used to study the correlation and degree of influence between variables.

Spearman's Rank analysis is a non-parametric statistical method used to measure the correlation between two variables. It assesses the monotonic relationship between variables by only calculating the rank order of the variables.

GDM is a spatial statistical analysis method mainly used to study the spatial correlation and influencing factors between geographical phenomena. It can identify the dominant role, interaction, and non-linear effects of different factors on the spatial distribution of geographical phenomena. It not only accounts for the rank order of the variables but also spatial information.

The results of Spearman's Rank analysis are shown in Table 8 and Table 9 below. Over the ECS, the correlation coefficient ρ between dependent a y variable (CER, COT, LWP) and an independent x variable (AOD, RH, LTS and PVV) are highest for AOD and following by PVV, LTS and RH. For CF and CTP, the correlation coefficient ρ is highest for PVV, followed by AOD, RH and LTS. The orders of correlation coefficient ρ are consistent with that of GDM q values (Table 5 in the revised manuscript). Over the YRD, for the CF the orders of correlation coefficient ρ are different from that of GDM q values (Table 6 in the revised manuscript). It shows that the correlation coefficient ρ is lowest for LTS but the GDM q value is highest for LTS. It may be attribute to that GDM not only accounts for the rank order of the variables as determined by the Spearman's Rank method but also spatial information.

Table 8. Statistics of Spearman's Rank analysis between x (AOD and meteorological conditions) and y (cloud parameters) over the ECS during 2008-2022. Statistically significant data points are indicated with * (p value < 0.01)**

Cloud parameter	AOD	RH	LTS	PVV
CER	-0.92***	0.61***	0.65***	-0.83***
COT	0.85***	-0.63***	-0.63***	0.83***
LWP	-0.85***	0.48***	0.59***	-0.71***
CF	0.65***	-0.46***	-0.23**	0.71***
CTP	-0.70***	0.73***	0.37***	-0.81***

Table 9. Statistics of Spearman's Rank analysis between x (AOD and meteorological conditions) and y (cloud parameters) over the YRD during 2008-2022. Statistically significant data points are indicated with * (p value < 0.01)

Cloud parameter	**AOD**	**RH**	**LTS**	**PVV**
CER	0.40***	-0.36***	-0.09	0.25**
COT	-0.76***	0.63***	-0.19	-0.42***
LWP	-0.35***	0.59***	-0.63***	-0.44***
CF	-0.49***	0.30***	0.26**	-0.32***
CTP	0.72***	-0.85***	0.48***	0.71***

22. L379 - The p-value for testing here is quoted as 0.01, but elsewhere it appears that 0.1 (a fairly lax criteria) is used.

Answer: Done. We have made unified standards that the p-value for testing here is quoted as 0.01 through the revised manuscript.

23. L422 - The high explanatory power of AOD for CF variations suggests that this method is not actually identifying causal relationships. While strong correlations between AOD and CF have been previously observed, they are likely due to aerosol humidification (Quaas et al, 2010), rather than an aerosol influence. It seems likely the same effect is being observed here, so care should be taken in the presentation of the results not to mis-attribute causality (unless applicable).

Answer: This study provides a general description of the sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters over YRD and ECS. Correlations between AOD and cloud parameters are found over the target regions, which can be attributed in part to the influence of general circulation. In general, there are many relations between the various parameters, both related to cloud microphysics and meteorology. It is not possible to completely separate meteorological influences from aerosol influences on clouds. This work can therefore only provide further evidence of the aerosol and meteorological effects on clouds and quantify the relative contributions and combined effects on clouds, but cannot quantify the absolute contributions with confidence. Thus, establishing cause and effect relationships between parameters is difficult and must be made with care.

We have added the following text in the Sect 4.6.1 (lines 647 to 666): "Tables 5 and 6 list q values for individual factors, together with p showing the absence of statistical significance in many cases, especially over the YRD, and often the explanatory power is not high when the significance is low. These data show that cloud parameters are dominated by aerosol effects over the ECS but meteorological influences on cloud parameters predominate over the YRD, as was also concluded from the analysis from "traditional" statistical methods presented in Section 4.5 and these conclusions are consistent with the results published by Andersen and Cermak (2015). Among the meteorological parameters, we also find that PVV (with highest q in the three meteorological parameters) predominantely influences cloud parameters over the ECS. Jones et al. (2009) and Jia et al. (2022) reported that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. In addition, we find that CTP is mainly affected by RH ($q = 0.74^{***}$) and PVV ($q = 0.56$) over the YRD, as suggested by Koren et al. (2010). Koren et al. reported that observed cloud top height correlates best with model pressure updraft velocity and relative humidity. To some extent, LTS influences CER ($q = 0.44^{***}$) and LWP ($q = 0.43^{***}$) over the ECS, while, in contrast, over the YRD LTS predominately influences CF ($q = 0.50^{***}$) and LWP ($q = 0.55^{***}$). Matsui et al. (2004) and Tan et al. (2017) reported that aerosol impact on CER is stronger in more dynamic environments that feature a lower LTS and argue that very high LTS environments dynamically suppress cloud droplet growth and reduce aci intensity. While strong correlations between AOD and cloud parameters have been previously observed, they are likely due to the swelling of aerosol particles in humid airmasses (Quaas et al, 2010), rather than an aerosol influence, which is in agreement with findings by, e.g., Myhre et al. (2007), Twohy et al. (2009) and Quaas et al. (2010).".

We have also added the following text in the Sect 4.6.2 (lines 652 to 655 and lines 695-699): "Among the meteorological parameters, we find that the combined effect of AOD and PVV predominately influences on cloud parameters over the ECS. The result is in accord with the finding of Jones et al. (2009) and Jia et al. (2022) that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions." and "The results from the GDM interaction detector analysis clearly show the enhancement of the interaction q-values over the q-values for the individual factors. In other words, the explanatory power of the combined effects of aerosol and a meteorological parameter is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters.".

24. L469 - After the introduction of the GDM, sections 4.5 and 4.6 appear to go back to more 'traditional' methods as used by previous paper. I am not sure I really see how these section support the paper in determining the cause of the different ACI values in these regions. It would be good to have a clearer link to the other work performed and how it supports the overall aim and conclusions of the paper.

Answer: In the revised version, we have moved Sections 4.5 (now 4.4.) and 4.6 (now 4.5) before Section 4.4 (now 4.6). Thus, we first discuss findings from "traditional" methods, followed by findings using the GDM. We have also added Section 5 (Discussion) and Section 6 (Conclusions) where we discuss the different findings using "traditional" methods and GDM, with more emphasis on the added value of GDM.

25. L601 - Could the authors be more specific on how this study will help improve model parametrisations?

Answer: Aerosol particles, acting as cloud condensation nuclei, affect the number and size of cloud droplets. The link between aerosol and the formation and properties of clouds could better simulate changes in cloud parameters. By comparing with observational data of aerosols and clouds, the model's ability to simulate changes in cloud parameters can be evaluated. Meteorological factors are key influencing parameters for the formation and evolution of clouds, and a more accurate description of the relative contribution of meteorological factors can improve the parameterization scheme of the model. Therefore, by more accurately simulating and predicting the impact of aerosols and meteorological parameters on clouds, parameterization schemes will be adjusted and improved, which further improve the simulation ability and accuracy of climate models for cloud parameter changes.

The text in the Conclusion has been reorganized as "By comparison with aerosol and cloud observations, the regional climate model's ability to simulate changes in cloud parameters can be evaluated. A more accurate description of the relative contribution of meteorological factors can improve the parameterization scheme of the model over eastern China." in the revision manuscript (lines 821-824).

References

We thanks referee#1 for providing these excellent references. They have been used in the manuscript and most of them have been quoted or summarized when appropriate. We have added some more references in the reference list of the revised manuscript.

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Response to Referee #2

This study uses correlation and the geographical detector method (GDM) to study relationships between aerosol optical depth (AOD), meteorological indicators, and cloud properties, contrasting a heavily polluted region in mainland China to a cleaner region of the Pacific Ocean influenced by transported pollution. The authors find different signs of the AOD-cloud effective radius relationships and find that AOD explains a very large fraction of variability in cloud properties, especially in the cleaner region.

The manuscript is well written, and the Figures and Tables illustrate the results and discussion well. But I have a mixed opinion about the study. On the one hand, there are innovative aspects, such as the application of the GDM to the aerosol-cloud problem. On the other hand, the study uses large-scale, time-averaged correlations of aerosols and clouds that have been shown to say little about aerosol-cloud interactions. And the impact of AOD on cloud variability that results from the GDM is so large that it would require a strong case to bring confidence in the method and results. On balance, I suggest major revisions to give the authors the chance to justify their results.

The authors thank Referee #2 for the valuable time spent on thorough reading our manuscript and providing expert views to guide us for improving the manuscript with the main and other comments. We have taken notice of all comments, listed below in black, and made many changes to the manuscript to address these, together with the comments from the other referees. We address each of your comments below and refer to our responses in the revised manuscript and provide line numbers and copy text in “quotes”.

To ensure that the data used only included single layer liquid clouds and nonprecipitating cases, the filtering criteria described by Saponaro et al. (2017) were applied. It is noted that all the figures have been updated throughout the revised manuscript.

Main comments:

1. Recent papers, and especially Gryspeerdt et al. (2023, 10.5194/acp-23-4115-2023) and Arola et al. (2022, 10.1038/s41467-022-34948-5), have seriously challenged the usefulness of correlating aerosol and cloud parameters as done in the present study. Cloud variability and retrieval errors are such that correlations between aerosol optical depth and cloud properties (CNDC, CER, LWP) can in fact be spurious. That means that a large fraction of past literature on aerosol-cloud interactions (including the studies cited lines 86-107) needs to be look at again critically. Attempts to minimise retrievals uncertainties (lines 172-175) will not address parts of the issues. I think the present study remains interesting (especially the GDM analysis), but the authors need to acknowledge the possibility that the correlations they find do not say much about aerosol-cloud interactions.

Answer: Gryspeerdt et al. (2023) discuss aci in terms of N_d rather than CER and emphasize the importance of the susceptibility β of N_d to aerosol. The main interest of their study is to determine RF_{aci} and the importance of accurate determination of β. The variation in β is responsible for much of the uncertainty in ERF_{aci} in climate models and β is central to the strength of cloud adjustments. Gryspeerdt et al. point out that uncertainties and differences occur with low aerosol conditions where satellite derived values of β are uncertain due to retrieval assumptions and separation of the (weak) aerosol signal from the surface reflectance. In high aerosol conditions, the (stronger) aerosol signal is relatively larger than the surface reflectance rendering more accurate aerosol retrieval in polluted conditions than in clean conditions. The larger uncertainty in clean condition reduces the correlation between CCN and the retrieved AOD due to regression dilution and thus reduces the magnitude of β in clean conditions. In the discussion, Gryspeerdt et al. argue that, for observational studies, the aerosol-N_d relationship is non-linear and the value of β determined for high aerosol conditions is not necessarily "a good guide" for β in low aerosol conditions. Furthermore, they argue that β in high conditions is more likely an underestimate.

In our study we used CER rather than N_d, for reasons discussed in the Introduction (see lines 177-183 in the revised manuscript): "It is noted that RF_{aci} is formulated in terms of N_d, whereas studies on the Twomey effects often use CER instead of N_d. CER is readily available as a satellite retrieval product, although in particular over land the reliability is questioned (Grandey and Stier, 2010), whereas N_d is derived from CER and the cloud optical thickness (COT) (e.g., Grandey and Stier, 2010; Arola et al., 2022). This implies that N_d is subject to the same retrieval errors as CER, including a possible relation between CER and LWP. The comparison of global maps of the sensitivities of CER and N_d to AOD by Grandey and Stier (2010) exhibits very similar patterns.", and we stratified by aerosol regime. We acknowledge the findings of Gryspeerdt et al. (2023) and possible consequences to our results with the following text added in Section 5, Discussion (lines 747-756):

"It is noticed that in recent papers (e.g., Gryspeerdt et al., 2023; Arola et al., 2022) the usefulness of correlating aerosol and cloud parameters has been seriously challenged because cloud variability and retrieval errors are such that correlations between AOD and cloud properties (N_d, CER, LWP) can be spurious. Gryspeerdt et al. (2023) discussed aci in terms of the susceptibility β of Nd to aerosol rather than the sensitivity S of CER to aerosol (see the discussion in the Introduction on the use of N_d vs CER), and the problem arises with low aerosol conditions due to larger aerosol retrieval uncertainty due to surface correction (larger surface effect on the radiance at the top of the atmosphere), which applies equally to β and S. In the current study we did not consider the lowest aerosol conditions by limiting the data to situations with AOD ≥ 0.1, as discussed in Section 4.2. Furthermore, we stratified the analysis for moderate (0.1 ≤ AOD < 0.3) and high (0.3 ≥ AOD) aerosol regimes, based on the data."

This text is followed by the discussion of the implications of the findings of Arola et al (2022) for our results (lines 757-777): "Arola et al. (2022) addressed the susceptibility of N_d to changes in aerosol and the adjustment of LWP (using satellite observations), and confounding factors, in particular co-variability of N_d and LWP induced by meteorological effects. They show how errors in the retrieved CER and COT or spatial heterogeneity in cloud fields influence the N_d - LWP relation. However, both N_d and LWP are not retrieved but derived from CER and COT. Using Eq. 1 and Eq. 2 in Arola et al. (2022), the N_d -LWP relationship can be shown to have a highly non-linear dependence on CER and thus it is no surprise that any error in CER strongly affects the relation between N_d and LWP. Their experiments, i.e. using smaller scales (5° x 5°) to reduce spatial meteorological variability, or using snapshots to remove meteorological variability in time, did not lead to a conclusion whether the N_d - LWP variability is due to spatial heterogeneity in the cloud fields or due to retrieval errors. The main message from this part of the study (using satellite data) by Arola et al. (2022) is "the spatial variability of CER introduces a bias which moreover becomes stronger in conditions where the CER values are lower on average". Experiments with simulated measurements show that "the main cause of the negative LWP vs N_d slopes is the error in CER". Arola et al. emphasize that the spatial cloud variability and retrieval errors in CER and COT are similar sources for negative bias in LWP adjustment and that these sources could not be separately assessed in their simulations.

The implication of the findings of Arola et al. (2022) on the adjustment of LWP for the results of the current study on the sensitivity of CER to aerosol (or CCN, using AOD as proxy) is that the assumption of constant LWP may be violated. This would affect the results presented in Section 4.3 where LWP was stratified and S was found to vary with LWP. In view of the LWP adjustment to changes in aerosol, the variation of CER sensitivity with LWP may be somewhat different from that reported in section 4.3."

2. The idea of using the GDM is interesting, but it is difficult to make physical sense of the results (Table 4 and 5). First, the q factors do not sum up to 1. What does that mean that AOD explains 87% of CER variability, while RH explains a further 36%? Then, the size of the q factors for AOD in Table 4, and to a lesser extent Table 5, stretches belief. If aerosols were that important in determining cloud properties, then estimating aerosol-cloud radiative forcing would have been very easy. Clearly, something goes wrong here. Is it perhaps that the four variables studied (AOD, RH, LTS, and PVV) are not independent? That the large q-values are simply a symptom of correlations caused by atmospheric circulation in the ECS and YRD? This is essentially what Figure 10 suggests. A strong discussion is needed to support the results.

Answer: A similar comment was made by Referee#1, i.e. that the q-values sum up to over 100% and about the interpretation of the q-values when the variables are not independent. Indeed, due to the interaction, the influence of a parameter may be strengthened or weakened and therefore the influence of different parameters need to be considered together. Therefore, our response is the same as that to Referee#1 Main point 1, while we also refer to the second part of our answer to your comment 3.

In statistics, the q-value is a measure used to evaluate the explanatory power of variables on the dependent variable. When multiple independent variables are considered separately, it is indeed possible for the sum of the q-values of multiple X variables to exceed 100%. When they are considered together, this is referred to as 'interaction q-value'. This situation is quite common and similar to the issue in multiple linear regression. The main reason for this is the presence of correlation among the X variables, indicating that these variables are not independent. Consequently, multiple independent variables may contribute to the dependent variable in a similar manner, leading to a sum of q-values over 100%.

To better explain this and clarify "interaction detector" and "interaction q-values", we have replaced the text below figure 2 (lines 354-374) with "The interaction detector can be used to test for the influence of interaction between different influencing factors, e.g., x1 and x2, on the dependent factor (y) and whether this interaction weakens or enhances the influence of each of x1 or x2 on the dependent variable, y, or whether they are independent in influencing y. For example, Figure 3(a) shows the spatial distribution of the dependent variable, y. The factors x1 and x2 both vary across the study region, but in different ways, and for each factor different sub-regions can be distinguished by application of the Jenks classification method described above to each factor separately. This is illustrated in Figures 3(b) and 3(c) where, as an example, three different sub-regions are considered for each factor. Usually, the dependent variable y is influenced by several different factors xi (Figure 3) and the combined effect of two or more factors may have a weaker or stronger influence on y than each of the individual factors. The q values for the influences of factors x1 and x2 on y, obtained from the application of the factor detector method (Eq. 2), may be represented as q (x1) and q (x2). Hence, a new spatial unit and subregions may be generated by overlaying the factor strata x1 and x2, written as x1∩x2, where ∩ denotes the interaction between factor strata x1 and x2 as illustrated in Figure 3(d).

Thus, the q value of the interaction of $x1 \cap x2$ may be obtained, represented as q ($x1 \cap x2$). Comparing the q value of the interaction of the pair of factors and the q value of each of the two individual factors, five categories of the interaction factor relationship can be considered which are summarized in Table 2. If $q(x1 \cap x2) > q(x1) + q(x2)$, this is referred to as a nonlinear enhancement of two variables. And if $q(x1 \cap x2) > Max[q(x1), q(x2)]$, this is referred to as a bilinear enhancement of two variables. The occurrence of nonlinear enhancement and bilinear enhancement are indicated with the q values in Table 2 and in the caption of Figure 7.”.

GDM is a spatial statistical analysis method aimed at studying the degree of influence and spatial patterns of different factors on the changes in geographic phenomena. In the analysis, we can simultaneously consider the interactions and impacts among multiple factors, thus revealing the relationships of synergistic changes. Therefore, geographic detector can encompass the analysis of synergistic changes.

We have added the following text in the Sect 4.6.1 (lines 647 to 666): “Tables 5 and 6 list q values for individual factors, together with p showing the absence of statistical significance in many cases, especially over the YRD, and often the explanatory power is not high when the significance is low. These data show that cloud parameters are dominated by aerosol effects over the ECS but meteorological influences on cloud parameters predominate over the YRD, as was also concluded from the analysis from “traditional” statistical methods presented in Section 4.5 and these conclusions are consistent with the results published by Andersen and Cermak (2015). Among the meteorological parameters, we also find that PVV (with highest q in the three meteorological parameters) predominantely influences cloud parameters over the ECS. Jones et al. (2009) and Jia et al. (2022) reported that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. In addition, we find that CTP is mainly affected by RH ($q = 0.74^{***}$) and PVV ($q = 0.56$) over the YRD, as suggested by Koren et al. (2010). Koren et al. reported that observed cloud top height correlates best with model pressure updraft velocity and relative humidity. To some extent, LTS influences CER ($q = 0.44^{***}$) and LWP ($q = 0.43^{***}$) over the ECS, while, in contrast, over the YRD LTS predominately influences CF ($q = 0.50^{***}$) and LWP ($q = 0.55^{***}$). Matsui et al. (2004) and Tan et al. (2017) reported that aerosol impact on CER is stronger in more dynamic environments that feature a lower LTS and argue that very high LTS environments dynamically suppress cloud droplet growth and reduce aci intensity. While strong correlations between AOD and cloud parameters have been previously observed, they are likely due to the swelling of aerosol particles in humid airmasses (Quaas et al, 2010), rather than an aerosol influence, which is in agreement with findings by, e.g., Myhre et al. (2007), Twohy et al. (2009) and Quaas et al. (2010).”

This study provides a general description of the sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters over the YRD and the ECS. Correlations between AOD and cloud parameters are found over the target regions, which can be attributed in part to the influence of general circulation. In general, there are many relations between the various parameters, both related to cloud microphysics and meteorology. Thus, establishing cause and effect relationships between parameters is difficult and must be made with care. It is not possible to completely separate meteorological influences from aerosol influences on clouds. This work can therefore only provide further evidence of the aerosol and meteorological effect on clouds and quantify the relative contributions and combined effects on clouds, but cannot quantify the absolute contributions with confidence.

In the current study, based on a regional scale of 9°x9°, the GDM method is used to explore the relative importance of various factors on cloud parameters and identify possible correlations between different factors. In the future, aerosol cloud interactions can be studied on smaller regional scale (<4°x4°) using higher resolution source data.

3. The GDM assumes that the spatial distributions of independent and dependent variables "should have evident similarities" [line 211]. But at what scale is that assumption true for aerosol-cloud interactions? One could expect the assumption to break down when going down to the scale of a cloud field because clouds evolve after their aerosol-influenced formation phase. Is that a problem?

Answer: Spatially-varying aerosol and cloud properties may contribute towards observed relationships between aerosol and cloud properties. This may affect the results of many of the aforementioned studies which analyze data on a relatively large regional scale. Aerosol type, cloud regime and synoptic regime may vary over large spatial scales. If data are analyzed for the region as a whole, false correlations may be introduced. Grandy and Stier (2010) suggested that for region sizes larger than 4°x4°, spurious spatial variations in retrieved cloud and aerosol properties can introduce widespread significant errors to calculation S. However, we can observe that at the regional scales of 8°x8° and 15°x15°, although significant errors are introduced, the spatial distribution patterns of S (the sensitivities of CER and N_d to AOD) look very similar, as shown in Figure 2 of Grandy and Stier (2010).

GDM is a spatial statistical analysis method aimed at studying the degree of influence and spatial patterns of different factors on the changes in geographic phenomena. In the analysis, we can simultaneously consider the interactions and impacts among multiple factors, thus revealing the relationships of synergistic changes. Therefore, geographic detector can encompass the analysis of synergistic changes.

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This study provides a general description of the sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters over the YRD and the ECS. Correlations between AOD and cloud parameters are found over the target regions, which can be attributed in part to the influence of general circulation. In general, there are many relations between the various parameters, both related to cloud microphysics and meteorology. Thus, establishing cause and effect relationships between parameters is difficult and must be made with care. It is not possible to completely separate meteorological influences from aerosol influences on clouds. This work can therefore only provide further evidence of the aerosol and meteorological effect on clouds and quantify the relative contributions and combined effects on clouds, but cannot quantify the absolute contributions with confidence.

In the current study, based on a regional scale of 9°x9°, the GDM method is used to explore the relative importance of various factors on cloud parameters and identify possible correlations between different factors. In the future, aerosol cloud interactions can be studied on smaller regional scale (<4°x4°) using higher resolution source data.

Other comments:

1. The name "ACI index" is vague. That quantity is really a sensitivity of cloud effective radius to changes in aerosol optical depth, in a similar way to beta_ln(N)-ln(tau) in Bellouin et al. (2020, doi: 10.1029/2019RG000660)

Answer: Following your suggestions below (in particular comment 5 referring to lines 55-78) we have changed the nomenclature and use sensitivity S (rather than β used in Bellouin et al. (2020)) and changed the title of Section 3.1 to "Sensitivity of cloud parameters to changes in aerosol concentrations" and used sensitivity throughout the manuscript. We also explained that we don't use cloud droplet number concentration (N_d) but cloud droplet effective radius (CER) and why we made this choice (see lines 175-186).

2. Lines 26 and 27: "significant" – is that in the statistical sense?

Answer: Yes, it is in the statistical sense and "significant" has been removed in order to make clear presentation.

3. Line 47: "in practice" in the atmospheric sciences. Other disciplines use the term more properly.

Answer: Thank you, we have changed "in practice" to "usually".

4. Lines 44-54: Those generalities on aerosols are not necessary, so that section could be shortened. In fact, the introduction could start directly from line 54: "Aerosol particles are important for climate…"

Answer: The manuscript discusses aerosol cloud interaction in contrasting regions as regards aerosol properties (high/low concentrations, composition). Therefore we feel that a short introduction on aerosols, their origin and their variability in space and time is appropriate. This short text also provides context for the description of the choice of study area (Section 2.1).

5. Lines 55-78: Note that many papers since Chapter 7 of the IPCC AR5 (Boucher et al. 2013) use the concept of aerosol-radiation and aerosol-cloud interactions and their respective adjustments (e.g., Bellouin et al. 2020, Quaas et al. 2022 10.5194/acp-22-12221-2022). The terms direct/1st indirect/2nd indirect remain in use in parts of the community, but it would be good to connect to the new terminology.

Answer: Thank you for this comment. Because the term "(in)direct" is still used quite often, also in recent publications, and in particular in the older literature, we had followed this terminology which is more common to us. However, we have of course also noticed the change and, although it was not easy to follow up on your comment, we have made an attempt and hope we have done it correctly, made no large mistakes, and done it everywhere where appropriate throughout the revised manuscript.

6. Lines 85-86: It should be said that using AOD as a proxy for aerosol concentrations when looking at aerosol-cloud interactions raises issues. See Section 6 of Bellouin et al. (2020).

Answer: Thank you for this comment. Indeed there are issues with the use of AOD and often AI (the product of AOD and the Ångström parameter, AE) is used. However there are also issues with AE from satellites and AE has even been withdrawn as a MODIS product from the more recent collections. The use of AOD in aci studies is discussed in the Introduction (lines 106-122): "In studies on S utilizing satellite data, which is the subject of the current study, the aerosol optical depth (AOD) is often used as a proxy for the aerosol concentration, which is justified by the correlation of AOD and CCN published by Andreae (2009). However, AOD is determined by all aerosol particles in the atmospheric column, including particles that do not act as CCN, depends on the relative humidity (RH) throughout the atmospheric column, does not provide information on chemical composition and may be influenced by aerosol in disconnected layers. The use of the Aerosol Index (AI), the product of AOD and the Ångström Exponent (AE; describing the spectral variation of AOD), is suggested as a better indicator of CCN because AE includes information on aerosol size (e.g., Nakajima et al., 2001). However, the AE is determined from AOD retrieved at two or more wavelengths and the evaluation of the results versus ground-based reference data shows the large uncertainty in AE. Therefore, in recent MODIS product Collections, AE is not provided over land (e.g., Levy et al., 2013; Kourtidis et al., 2015). AE is also not well-defined for low AOD for which uncertainty is largest (Bellouin et al., 2020; Gryspeerdt et al., 2023). The issues associated with using AOD or AI as proxy for CCN were discussed by, among others, Rosenfeld et al. (2014) who do not recommend the use of AI while also concluding that no better proxy is available. Therefore, in this study, AOD is used as a proxy for CCN to study S. It is noted that in other studies, e.g., Jia et al., 2022, both AOD and AI have been used and the results show similar behaviour.".

7. Line 168: Andreae (2009) is often cited as justification for using AOD for looking at aerosol-cloud interactions, but ironically its Figure 1 shows that the correlation only exists across aerosol regimes. For a given regime (as done in the present study) there is essentially no correlation. I could not see why Kourtidis et al. (2015) justifies the use of AOD, but I may have missed it.

Answer: See our response to your comment 6 as regards using AOD as aerosol proxy. Specifically to this comment 7: Andreae (2009) plots AOD vs CCN for 4 aerosol types, which happen to be separated in two groups for low and high CCN and indeed the correlation was evaluated across these 4 aerosol regimes. However, it is noted that the number of data pairs is scarce but within each aerosol type the AOD increases with increasing CCN. This AOD-CCN relation may however vary between aerosol types (as would be expected for aerosol types with different composition and thus also hygroscopic properties), but the number of data points is too small to derive different relationships for different aerosol types. Furthermore, the aerosol types over each of the two study regions varies with season (seasonal emissions like desert dust in spring, biomass burning from different sources, domestic heating in winter) and large scale meteorological condition resulting in different transport pathways in different seasons.

We referenced Kourtidis et al. (2015) because these authors justify the use of AOD instead of AE based on personal communication with Lorraine Remer, as mentioned in our response to your comment 6.

8. Lines 169-170: That assumes that cloud contamination has a lesser impact on smaller AODs. Is that true?

In aerosol retrieval, cloud screening is a major source for over-estimation of the AOD, in particular in the vicinity of clouds which is important for aci. The impact of undetected clouds depends on the COD, and may be small (in an absolute sense) for thin Cirrus clouds, but also discrimination between high AOD and clouds is often a problem. The removal of cloud-contaminated pixels is not straightforward. A post-processing method shows the effect of removal of residual clouds on the AOD in both relatively clean and polluted areas (doi:10.5194/amt-10-491-2017). Alternative methods have been proposed such as setting a threshold for AOD < 0.6 proposed by Brendan et al. (2006) who used the MOD06 Collection 04 cloud product. Brendan et al. (2006) conclude with "The cloud masking technique in the recently updated Collection 05 cloud retrieval algorithm has been improved, and the Collection 05 cloud products available in the near future will largely eliminate the aerosol contamination effect". Christenson et al. (ACP 2017) used MOD06 C6 data (1km x1km) and reported that "large aerosol optical depths remain in the MODIS-observed pixels near cloud edges, due primarily to 3-D effects (Varnái and Marshak, 2009) and the swelling of aerosols by higher relative humidity." And "Varnái and Marshak (2009) also noted that beyond 15 km contamination effects were minimized in MODIS data (1km x1km)." Therefore Christensen et al. only used data pairs beyond the 15 km length scale in their aci study.

In our study we use MODIS L3 collection 6.1 with a spatial resolution of 1°x1°. Comparisons with surface-based sun photometer data revealed that Collection 6 should improve upon Collection 5, and overall, 69.4% of MODIS Collection 6 AOD fell within an expected uncertainty of ± (0.05 + 15%) (Levy et al., 2013; Tan et al., 2017). In this study, to eliminate 1° by 1° scenes in which the aerosol distribution is heterogeneous, retrievals with a standard deviation higher than the mean values are discarded (Saponaro et al., 2017; Jia et al., 2022). In addition, many previous researches do not set a threshold of AOD when using MODIS L3 C6 data (Grandey and Stier, 2010; Tang et al., 2014; Saponaro et al., 2017; Tan et al., 2017; Ma et al., 2018; Jia et al., 2022). Based on these findings, we used the larger threshold of 1.5.

We have added the following text to Section 2.2 (lines 252-260): "The choice of this threshold, rather than 0.6 used by Brendan et al. (2006), who used MOD06 Collection 04 products, is based on reports by Christenson et al. (2017) and (Varnái and Marshak, 2009). Christenson et al. (2017) used MOD06 C6 data (1km x1km) and reported that "large aerosol optical depths remain in the MODIS-observed pixels near cloud edges, due primarily to 3-D effects (Varnái and Marshak, 2009) and the swelling of aerosols by higher relative humidity." Varnái and Marshak (2009) noted that beyond 15 km contamination effects were minimized in MODIS data (1km x1km). Furthermore, we discarded scenes (1° by 1°) in which the aerosol distribution is heterogeneous, i.e. with a standard deviation higher than the mean value (Saponaro et al., 2017; Jia et al., 2022)."

9. Line 207: "intermittently" Probably not the correct word. Interchangeably?

Answer: Thank you for this suggestion, we have changed "intermittently" to "Interchangeably"

10. Line 231: Does q sum up to 1 for all factors considered? Is it also able to quantify an unexplained fraction that could suggest the need for more factors?

Answer: This comment is similar to a comment made by Referee#1 and was addressed in our response to your comment 2. Because the contribution of each independent variable (each factor) is calculated separately according to Eq. (2), the contributions of some factors (if needed) that are not considered can also be calculated separately according to Eq. (2).

11. Figure 3: What does that Figure tell the reader? It's impossible to say from its caption or from lines 241-245. The discussion needs to cover each of the panels in turn.

Answer: To better explain the Figure and clarify "interaction detector" and "interaction q-values", we have replaced the text below figure 2 (see lines 354-374 in the revised manuscript) with "The interaction detector can be used to test for the influence of interaction between different influencing factors, e.g., x1 and x2, on the dependent factor (y) and whether this interaction weakens or enhances the influence of each of x1 or x2 on the dependent variable, y, or whether they are independent in influencing y. For example, Figure 3(a) shows the spatial distribution of the dependent variable, y. The factors x1 and x2 both vary across the study region, but in different ways, and for each factor different sub-regions can be distinguished by application of the Jenks classification method described above to each factor separately. This is illustrated in Figures 3(b) and 3(c) where, as an example, three different sub-regions are considered for each factor. Usually, the dependent variable y is influenced by several different factors xi (Figure 3) and the combined effect of two or more factors may have a weaker or stronger influence on y than each of the individual factors. The q values for the influences of factors x1 and x2 on y, obtained from the application of the factor detector method (Eq. 2), may be represented as q (x1) and q (x2). Hence, a new spatial unit and subregions may be generated by overlaying the factor strata x1 and x2, written as $x1 \cap x2$, where $\cap$ denotes the interaction between factor strata x1 and x2 as illustrated in Figure 3(d). Thus, the q value of the interaction of $x1 \cap x2$ may be obtained, represented as $q\ (x1 \cap x2)$. Comparing the q value of the interaction of the pair of factors and the q value of each of the two individual factors, five categories of the interaction factor relationship can be considered which are summarized in Table 2. If $q(x1 \cap x2) > q(x1) + q(x2)$, this is referred to as a nonlinear enhancement of two variables. And if $q(x1 \cap x2) > Max[q(x1), q(x2)]$, this is referred to as a bilinear enhancement of two variables. The occurrence of nonlinear enhancement and bilinear enhancement are indicated with the q values in Table 2 and in the caption of Figure 7.".

12. Lines 251-252: "for several different purposes". Give examples based on the papers cited.

Answer: Examples based on the GDM have been added in the revised manuscript below Table 2 (lines 383-387): "The geographical detector method has been used to detect influencing factors for several different purposes (e.g., Wang et al., 2018; Zhang and Zhao, 2018; Zhou et al., 2018). For example, the GDM was used to detect the influence of annual and seasonal factors on the spatial-temporal characteristics of surface water quality (Wang et al., 2018). Other examples are the application of the GDM to examine factors influencing regional energy-related carbon emissions (Zhang and Zhao, 2018) and to examine effects of socioeconomic development on fine particulate matter (PM2.5) in China (Zhou et al., 2018)."

13. Line 265: "averaged over the years 2008-2022". Does the study use 14-year averaged distributions, or a less dramatic averaging (e.g., multi-annual monthly means)? How can correlation of distributions averaged over such a long period still inform about the physical correlation between clouds and aerosols?

Answer: In Figure 4 we provide and overview of the data as averages over the whole study period (2008-2022). However, in the text we indicate that individual data pairs are used in the research, e.g., in Section 3.2. (lines 338-339) "In this study, multi-years of mean values of influencing factors (x) and dependent factors (y) were calculated for each raster grid" and in Section 4.2. (lines 429-431) "To investigate S, we used correlated data pairs for 15 years and the data was binned in AOD intervals with a bin width of 0.02, and the CER data in each AOD bin were averaged."

The use of multi-year averages is not uncommon in aci studies, e.g. Ma et al. (GRL 2018) use 2003-2016. Such large samples allow for large numbers of data pairs. However, with hindsight we agree that 15 years is very long. We have thought about shorter periods and realized that, in principle, periods were included when the AOD was at its maximum (2008-2014) and when the AOD was decreasing in response to implementation of emission reduction policy. We therefore split the data sets for these 2 periods and plotted CER vs AOD, see the Figures 1 and 2 below. We noticed that over the ECS there was not a significant difference between the CER/AOD relations during these two periods. However over the YRD, for the high AOD period, CER clearly decreased with increasing AOD for 0.1<AOD<0.3 and for larger AOD the CER increased with R=0.87. For the second period, however, there was no clear correlation between CER and AOD for both AOD intervals. The data also show that over the YRD the CER for AOD>0.3 increased to larger values during the first period than during the second period. We did not look for explanations of this difference, possibly the aerosol properties changed in response to emission reduction, or confounding meteorological factors played a role.

We also looked for shorter periods, considering each year between 2008 and 2022. The results show similar behavior for each year over both study areas with interannual variations between the fits, and thus the value of S. However, the statistical significance is low (large p) due to the small number of samples.

These findings were briefly summarized in the discussion (lines 733-746): "These results were obtained using data from a period of 15 years. During this period, the aerosol properties changed in response to expanding economy, resulting in the increase of the AOD until 2007, and the implementation of emission reduction policy resulting in the decrease of the AOD from 2014 which flattened from about 2018 (de Leeuw et al., 2021; 2022; 2023). To account for these changes, the sensitivity S was determined for the periods 2008-2014 and 2014-2022, without stratification for LWP (see Figures S1 and S2 in the Supplementary). The results for the ECS show no significant difference between the CER-AOD relations during these two periods. Over the YRD, however, the data for 2008-2014 show a clear decrease of CER with increasing AOD for 0.1<AOD<0.3 and for larger AOD the CER increased, with a statistical significant correlation (R=0.87) and S=0.10 as compared to S=0.08 for the whole period. In contrast, the data for 2014-2022 show no clear correlation between CER and AOD for both AOD intervals over the YRD. A similar exercise for shorter periods, i.e. for each year between 2008 and 2022, show similar behavior as for the whole period 2008-2022, over both study areas, with interannual variations of the value of S. However, the statistical significance is low (large p) due to the small number of data samples in each year."

In the GDM, the y data are recorded in a raster grid, over a total study area of 9°x9°, as illustrated in Figure 3 (Figure 2 in the revised manuscript). The data in the raster grid is transformed into dot files, each dot containing a value for y and for one of the influencing parameters x. The dependent (y) and influencing (x) parameters are separated into 2 layers with the same grid. As the resolution of MYD08 data used in this study is 1°x1°, the data transformed into dot files is based on raster grid 1°x1°. Thus, 15-year averaged distributions of clouds (y, 5 layers) and aerosols/meteorological conditions (x, 4 layers) are used as input in the GDM. This is specified in the text on page 13 of the revised manuscript (see lines 338-341): "In this study, multi-years of mean values of influencing factors (x) and dependent factors (y) were calculated for each raster grid. Then, we classified the influencing factors (e.g. AOD and meteorological parameters) into 5 sub-regions by the Jenks natural breaks classification method (Brewer and Pickle, 2002).".

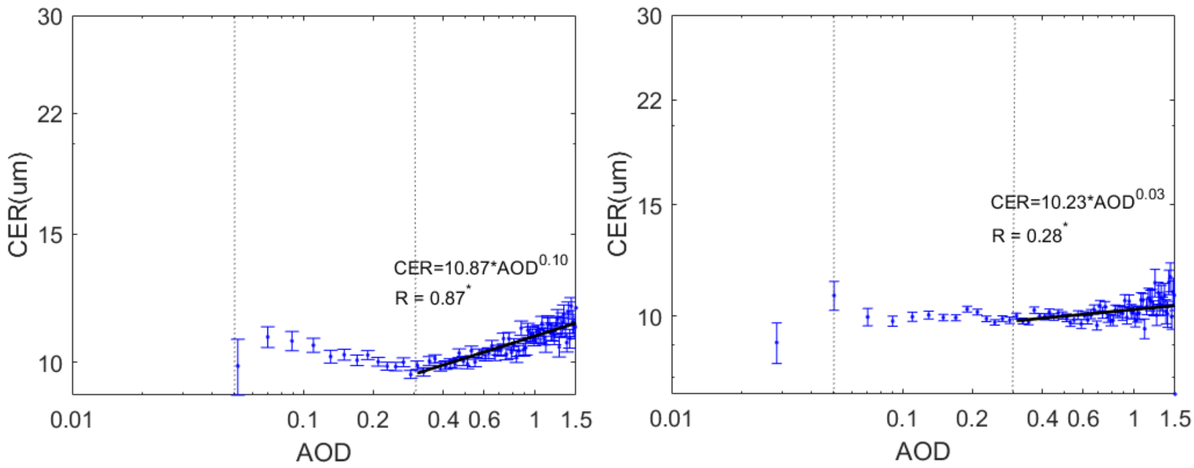

Figure 1. CER vs AOD over the YRD for the periods 2008-2014 (left) and 2015-2022 (right).

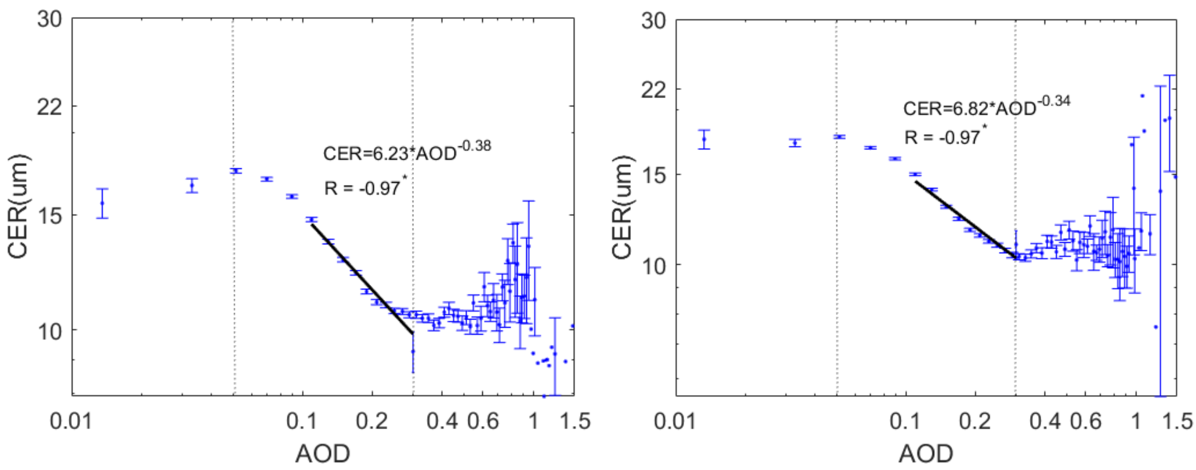

Figure 2. CER vs AOD over the ECS for the periods 2008-2014 (left) and 2015-2022 (right).

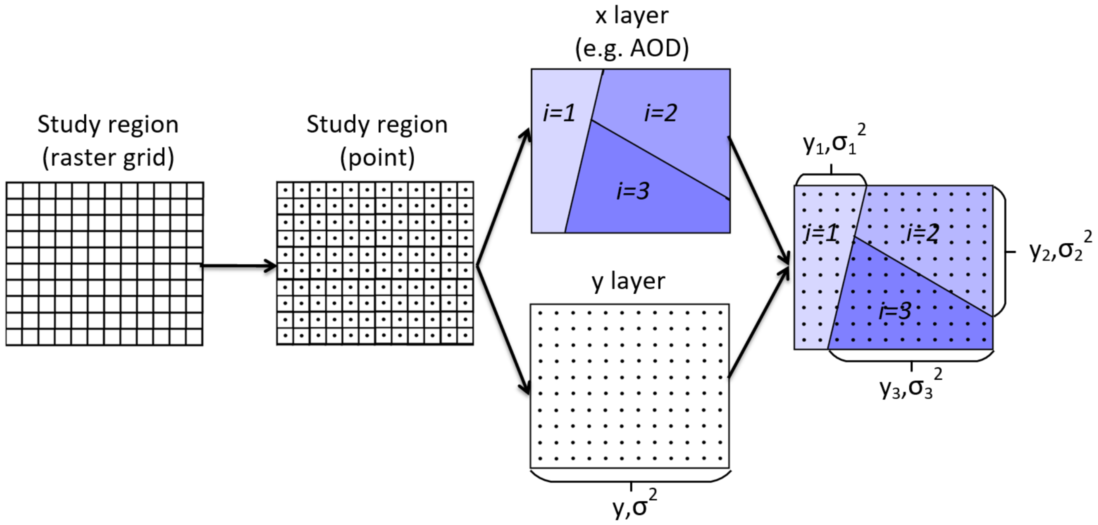

Figure 3. The principle of the geographical detector method. See text for explanation.

14. Lines 285-286: Or it could be due to meteorological factors.

Answer: In this section we describe the observations based on Figure 4 in the revised manuscript. We note that over ocean the aerosol properties are different than over land and certainly sea spray aerosol is abundant, while over land other aerosol types dominate. In the next Section we notice that the sensitivity of CER to AOD is larger over ocean than over land, which confirms the observations in lines 285-286 (now lines 421-422 in the revised manuscript). Certainly, meteorological factors influence the aci too and the interactive q-factors presented and discussed in Section 4.6.2 show that the combined effect has a larger influence than the effect of one factor alone. However, we prefer to structure the paper and go step by step through the different aspects. To explain this, we have added the following text above Figure 4 (lines 422-424): "The influence of different factors on the sensitivity of cloud parameters to aerosol and the adjustments are discussed in the following sections, based on both statistical methods and the application of the GDM."

15. Line 373: Missing word: "in the range of"

Answer: Thank you for this comment: corrected.

Response to Referee #3

This study investigates the aerosol and meteorological parameters on warm clouds using satellite measurements. The authors focus on the period of 2008-2022 over the two contrasting regions over eastern China, i.e. Yangtze River Delta, a heavily polluted region in eastern China, and the East China Sea with a relatively clean atmosphere. The interaction between AOD and CER has been investigated by considering different AOD and LWP regimes in the both two different aerosol regimes. A new method (geographical detector method) was applied to explore the relative importance of AOD and meteorological parameters on cloud properties. The content of this manuscript is highly relevant to ACP readers. In general, the manuscript is well organized, and the analysis conducted is quite comprehensive. Based on the overall quality, it is recommended that the manuscript be considered for publication if the specific comments provided are addressed.

The authors thank Referee #3 for the valuable time spent on thorough reading our manuscript and providing expert views to guide us for improving the manuscript with the specific comments and a reference. We have taken notice of all comments, listed below in black, and made many changes to the manuscript to address these, together with the comments from the other referees. We address each of your comments below and refer to our responses in the revised manuscript and provide line numbers and copy text in “quotes”.

To ensure that the data used only included single layer liquid clouds and nonprecipitating cases, the filtering criteria described by Saponaro et al. (2017) were applied. It is noted that all the figures have been updated throughout the revised manuscript.

Specific comments

1. Abstract: it would be beneficial if the authors emphasized the overall significance or implications of their study at the end of the abstract.

Answer: We have substantially revised the abstract and added to following sentence upfront, to provide the overall picture “The sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters has in most previous aerosol-cloud interaction (aci) studies been addressed using traditional statistical methods. In the current study, relationships between cloud droplet effective radius (CER) and aerosol optical depth (AOD, used as a proxy for cloud condensation nuclei, CCN), i.e. the sensitivity (S) of CER to AOD, is investigated with different constraints of AOD and cloud liquid water path (LWP). In addition to traditional statistical methods, the geographical detector method (GDM) has been applied to quantify the relative importance of the effects of aerosol and meteorological parameters, and their interaction, on S.”. In addition, many other changes were made to the abstract in “track changes”.

2. In order to provide a more comprehensive analysis, it would be beneficial for the authors to compare the results obtained in this study with findings from other regions around the world. By doing so, they can examine the unique aerosol effects on clouds in the specific target region.

Answer: In the revised text, the results are compared with many other findings. We have added the following text in the Sect 4.3 (lines 528 to 534): “The variation of S with changes in LWP indicates that the condition of constant LWP is not truly satisfied: if the data would be stratified according to smaller LWP intervals (quasi-constant LWP, Ma et al., 2018), S would likely vary more smoothly with LWP. As mentioned in the Introduction, LWP is not directly retrieved but calculated form CER and COT and thus also the calculation of S is to some extend affected by LWP. We further note the results by Ma et al. (2018), i.e. the slope of CER versus AI (comparable to S in this paper) varies little with LWP, with positive values over land and negative values over ocean and thus behaves similar to the data in Table 3 for YRD and ECS."

We have also added the following text in the Sect 4.3 (lines 647 to 666): "Tables 5 and 6 list q values for individual factors, together with p showing the absence of statistical significance in many cases, especially over the YRD, and often the explanatory power is not high when the significance is low. These data show that cloud parameters are dominated by aerosol effects over the ECS but meteorological influences on cloud parameters predominate over the YRD, as was also concluded from the analysis from "traditional" statistical methods presented in Section 4.5 and these conclusions are consistent with the results published by Andersen and Cermak (2015). Among the meteorological parameters, we also find that PVV (with highest q in the three meteorological parameters) predominantely influences cloud parameters over the ECS. Jones et al. (2009) and Jia et al. (2022) reported that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. In addition, we find that CTP is mainly affected by RH ($q = 0.74^{***}$) and PVV ($q = 0.56$) over the YRD, as suggested by Koren et al. (2010). Koren et al. reported that observed cloud top height correlates best with model pressure updraft velocity and relative humidity. To some extent, LTS influences CER ($q = 0.44^{***}$) and LWP ($q = 0.43^{***}$) over the ECS, while, in contrast, over the YRD LTS predominately influences CF ($q = 0.50^{***}$) and LWP ($q = 0.55^{***}$). Matsui et al. (2004) and Tan et al. (2017) reported that aerosol impact on CER is stronger in more dynamic environments that feature a lower LTS and argue that very high LTS environments dynamically suppress cloud droplet growth and reduce aci intensity. While strong correlations between AOD and cloud parameters have been previously observed, they are likely due to the swelling of aerosol particles in humid airmasses (Quaas et al, 2010), rather than an aerosol influence, which is in agreement with findings by, e.g., Myhre et al. (2007), Twohy et al. (2009) and Quaas et al. (2010)."

We have also added the following text in the Discussion (lines 747 to 756): "It is noticed that in recent papers (e.g., Gryspeerdt et al., 2023; Arola et al., 2022) the usefulness of correlating aerosol and cloud parameters has been seriously challenged because cloud variability and retrieval errors are such that correlations between AOD and cloud properties (N_d, CER, LWP) can be spurious. Gryspeerdt et al. (2023) discussed aci in terms of the susceptibility β of Nd to aerosol rather than the sensitivity S of CER to aerosol (see the discussion in the Introduction on the use of N_d vs CER), and the problem arises with low aerosol conditions due to larger aerosol retrieval uncertainty due to surface correction (larger surface effect on the radiance at the top of the atmosphere), which applies equally to β and S. In the current study we did not consider the lowest aerosol conditions by limiting the data to situations with AOD ≥ 0.1, as discussed in Section 4.2. Furthermore, we stratified the analysis for moderate ($0.1 \leq$ AOD < 0.3) and high ($0.3 \geq$ AOD) aerosol regimes, based on the data."

This text is followed by the discussion of the implications of the findings of Arola et al (2022) for our results (lines 757-777): "Arola et al. (2022) addressed the susceptibility of N_d to changes in aerosol and the adjustment of LWP (using satellite observations), and confounding factors, in particular co-variability of N_d and LWP induced by meteorological effects. They show how errors in the retrieved CER and COT or spatial heterogeneity in cloud fields influence the N_d -LWP relation. However, both N_d and LWP are not retrieved but derived from CER and COT. Using Eq. 1 and Eq. 2 in Arola et al. (2022), the N_d -LWP relationship can be shown to have a highly non-linear dependence on CER and thus it is no surprise that any error in CER strongly affects the relation between N_d and LWP. Their experiments, i.e. using smaller scales (5° x 5°) to reduce spatial meteorological variability, or using snapshots to remove meteorological variability in time, did not lead to a conclusion whether the N_d - LWP variability is due to spatial heterogeneity in the cloud fields or due to retrieval errors. The main message from this part of the study (using satellite data) by Arola et al. (2022) is "the spatial variability of CER introduces a bias which moreover becomes stronger in conditions where the CER values are lower on average". Experiments with simulated measurements show that "the main cause of the negative LWP vs N_d slopes is the error in CER". Arola et al. emphasize that the spatial cloud variability and retrieval errors in CER and COT are similar sources for negative bias in LWP adjustment and that these sources could not be separately assessed in their simulations. The implication of the findings of Arola et al. (2022) on the adjustment of LWP for the results of the current study on the sensitivity of CER to aerosol (or CCN, using AOD as proxy) is that the assumption of constant LWP may be violated. This would affect the results presented in Section 4.3 where LWP was stratified and S was found to vary with LWP. In view of the LWP adjustment to changes in aerosol, the variation of CER sensitivity with LWP may be somewhat different from that reported in section 4.3."

3. Page 5，line 146：add "." in the end.

Thank you: done

4. Page 6，line 150：change "Eastern China Sea (ECS) area (20°N-28°N, 126°E-134°E)" to "Eastern China Sea area (ECS, 20°N-28°N, 126°E-134°E)"

Thank you: done

5. Page 7，line 191：change "(cloud optical thickness, cloud droplet effective radius, etc.)" to "(COT, CER, etc.)".

Thank you: done

6. Page 8, line 202: change "Where re represents the cloud droplet effective radius (CER)" to "Where re represents the CER".

Thank you: done

7. Page 11, line 276: it suggests to define the acronyms about "NW" and "SW".

Thank you. We feel that the use of wind directions is common in geographical descriptions of spatial variation and since we refer here to the maps in Fig. 4, it is not necessary to define the abbreviations for the wind directions. However, after reading the relevant text again, we noticed that we have used the full names for other wind directions (like south) and therefore decided to write them in full throughout the manuscript, i.e. we replace NW with northwest etc.

8. Page 14, line 344: remove right parenthesis.

Thank you: done

9. Page 15, line 365: replace "for the YRD" with "over the YRD".

Thank you: done

10. Page 21, line 501: add right parenthesis after “by Liu et al., (2017”.

Thank you: done

11. Page 22, line 516: change “for three different LWP intervals” to “for five different LWP intervals”.

Thank you: done

12. Page 16, line 377-380: The statistically significance is used through the manuscript, so it suggests to describe at the first place in the manuscript.

Answer: We have added the following text in the Sect 3.1 (lines 307 to 310): “The significance of these relations is determined by using the student’s t test, i.e. the results are statistically significant when the p value is smaller than 0.01, where p is defined as the probability of obtaining a result equal to or “more extreme” than what was actually observed.”.

References

Ma, X., Jia, H., Yu, F., & Quaas, J. (2018). Opposite aerosol index-cloud droplet effective radius correlations over major industrial regions and their adjacent oceans. Geophysical Research Letters, 45, 5771–5778. https://doi.org/10.1029/2018GL077562

Opposite effects of aerosols and meteorological parameters on warm clouds in two contrasting regions over eastern China

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Abstract. ~~Most of previous studies addressed T~~the sensitivity (S) of cloud parameters to the influence of different aerosol and meteorological parameters ~~on the sensitivity of cloud parameters to aerosol indirect effect~~ has in most previous aerosol-cloud interaction (aci) studies been addressed using traditional statistical methods. In the current study, relationships between cloud droplet effective radius (CER) and aerosol optical depth (AOD, used as a proxy for ~~aerosol concentration~~cloud condensation nuclei, CCN~~, concentrations~~), ~~characterized by~~i.e. the sensitivity (S) of CER to AOD~~ (S)~~, is investigated with~~were constructed for~~ different constraints of AOD and cloud liquid water path (LWP). In addition to traditional statistical methods, ~~T~~the geographical detector method (GDM) ~~is~~is applied in this study to quantify the relative importance of the effects of aerosol and meteorological parameters, and their interaction, on S. ~~Aerosol and cloud properties retrieved from t~~The Moderate Resolution Imaging Spectroradiometer (MODIS) ~~on-board the Aqua satellite~~C6 L3 data and ~~the~~ European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-5 reanalysis data, for the period from 2008 to 2022, were used to investigate ~~aerosol-cloud interaction (ACI~~aciACIaci~~)~~ over eastern China~~, using aerosol optical depth (AOD) as a proxy for the aerosol concentration, during a period of 15 years (2008-2022)~~. Two contrasting areas were selected: the heavily polluted Yangtze River Delta (YRD) and a relatively clean area over the East China Sea (ECS). Linear regression analysis shows that ~~the opposite~~ sensitivity (S) of ~~behaviour of the cloud droplet effective radius (CER)~~ to ~~and AOD relationship in the two different aerosol regimes.~~ CER

decreases with the increase of AOD (negative S) in the moderately polluted atmosphere (0.1<AOD<0.3) over the ECS, whereas, in contrast,~~, in agreement with the Twomey effect. However,~~ ~~but~~ CER increases with increasing AOD (positive S) in the polluted atmosphere (AOD>0.3) over the YRD~~, CER increases with increasing AOD (positive S)~~. Evaluation of S~~the ACI index (here defined as the change in CER as a function of AOD)~~ as function of the ~~cloud liquid water path (~~LWP~~)~~ shows that in the moderately polluted atmosphere over the ECS, S~~the ACI index~~ is ~~significant and positive~~ negative in the LWP interval [40 g m^{-2}, 200 g m^{-2}], and the sensitivity of CER to AOD is ~~increases~~ substantially stronger as ~~with~~ LWP ~~increasing LWP~~is larger. In contrast, in the polluted atmosphere over the YRD, S ~~the ACI index~~ is ~~significant and negative~~ positive in the LWP interval [0 g m^{-2}, 120 g m^{-2}] and does not change notably as function of LWP in this interval. The study further shows that over the ECS the CER is larger for higher LTS and RH but lower for higher PVV. Over the YRD, there is no significant influence of LTS on the relationship between CER and AOD. ~~Furthermore, t~~The GDM has been used as an independent method to~~To further~~ analyse the sensitivity of cloud parameters to ~~influence of~~ AOD and meteorological ~~conditions~~parameters (relative humidity, RH~~,~~; lower tropospheric stability, LTS~~,~~; and pressure vertical velocity, PVV ~~on cloud parameters~~). The GDM has also been used to analyse the effects of interactions between two parameters and thus obtain information on confounding meteorological effects on the aci. ~~,, the geographical detector method (GDM) has been used. The results show that all factors have a significant influence on the cloud parameters~~ ~~O~~over the ECS, cloud parameters are sensitive to ~~all~~almost ~~of~~all parameters considered~~,~~ except for cloud top pressure (CTP), ~~but~~ and the sensitivity to ~~influence of~~ AOD is larger than that ~~of~~ to any of the meteorological factors. Among the meteorological factors, ~~lower tropospheric stability (LTS) has the largest influence on~~ the cloud parameters are most sensitive to ~~LTS~~PVV and least sensitive to ~~relative humidity (~~RH~~) the smallest~~. Over the YRD, the explanatory power of the sensitivity of cloud parameters to ~~effect of~~ AOD and meteorological parameters ~~on cloud parameters~~ is much smaller than over the ECS, except for RH which has a statistically significant influence on CTP and can explain ~~65~~74% of the variation of CTP. The results from the GDM analysis show that the explanatory power of the combined effects of aerosol and a meteorological parameter is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters.~~The combined effect of meteorological factors and AOD on cloud parameters enhances the explanatory power over the effect~~

~~of individual parameters. The study further shows that over the ECS the effect of RH and LTS on the CER/AOD relationship is opposite to that of pressure vertical velocity (PVV). Over the YRD, The CER is larger in unstable atmospheric conditions than in stable conditions, irrespective of the AOD and the CER is much larger in high relative humidity conditions than in low relative humidity conditions.~~

Key words: AOD, Cloud parameters, LWP, Geographical detector method, Confounding effects, MODIS, East China

1 Introduction

The atmosphere is primarily composed of gases, i.e. nitrogen, oxygen and several noble gases, as well as a wide variety of trace gases that occur in relatively small and highly variable amounts. In addition, liquid and solid particles are suspended in the atmosphere. The suspension of solid and liquid particles in the gaseous medium is technically defined as an aerosol, but ~~in practice~~usually the term aerosol refers to the particulate component only (Seinfeld and Pandis, 1998). The aerosol particles originate from a large variation of both direct and indirect sources. ~~–and t~~The concentrations and ~~in~~ chemical and physical properties of aerosol particles change under the influence of a variety of atmospheric processes and ~~, which~~ thus are variable in space and time. The residence time of tropospheric aerosol particles varies from hours to weeks (Bellouin et al., 2020), depending on particle size and atmospheric conditions. Directly emitted aerosol types include, e.g., sea spray, dust, smoke, volcanic ash, pollen etc. ~~The indirect~~Secondary formation of aerosol particles occurs through nucleation and subsequent growth by physical and chemical processes such as condensation, coagulation and multiphase chemical reactions on the particle surface, involving precursor gases such as sulphur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3), volatile organic compounds (VOCs), etc.

Aerosol particles are important for climate, air quality and heterogenous chemical processes. Aerosol particles ~~exert a direct e~~affect ~~on~~ climate by their interaction with radiation (aerosol radiation interaction, ari) which exerts a radiative forcing (RF_{ari}) on the Earth'-energy budget which results in rapid adjustments of global mean atmospheric quantities such as temperature. The sign and strength of radiative forcing (RF) due to ari ~~–~~(RF_{ari}) vary with environmental parameters (Bellouin et al., 2020). In particular, aerosol particles~~by~~ scatter~~ing~~ incoming solar radiation back into space, but the effect of RF_{ari} depends on the brightness of the aerosol with respect to that of the underlying surface. The scattering of (bright) aerosol over a darker surface ~~which~~ results in cooling and reduction of the warming effect of greenhouse gases (GHG). ~~, while~~In contrast, the interaction of absorbing aerosol particles with solar radiation may result in local heating and thus reinforce the GHG effect and influence meteorological processes.

Aerosol particles can act as cloud condensation nuclei (CCN, in liquid clouds) or ice nucleating particles~~i~~ (INP, in ice clouds), depending on their chemical composition and size. When CCN~~; when they~~ are activated they can ~~exert an indirect effect on climate by~~ modify ~~changing the~~ cloud microphysical properties and precipitation and thus indirectly influence the Earth's radiative budget (aerosol-cloud interactions, aci) (Tao et al., 2012; Fan et al., 2016; Rosenfeld et al., 2019; Rao and Dey, 2020; Bellouin et al., 2020). An increase in CCN concentrations leads to an increase in the number of cloud droplets (N_d) and, if~~The first indirect effect of aerosols, often referred to as the "Twomey" effect (Twomey, 1977; Matheson et al., 2005; Koren et al., 2005; Meskhidze and Nenes, 2010; Costantino et al., 2010; 2013), describes the effect of the increase of the number of aerosol particles when~~ the cloud liquid water path (LWP) remains unchanged, ~~which results in the increase in the number of cloud droplets and~~ the decrease of the cloud droplet effective radius (CER). The smaller CER in turn results in the enhanced reflection of solar radiation and thus cloud albedo and enhanced RF due to aci (RF_{aci}). This effect of the increase of the number of aerosol particles on cloud properties at constant LWP is often referred to as the "Twomey" effect (Twomey, 1977; Feingold, et al., 2001; Matheson et al., 2005; Koren et al., 2005; Meskhidze and Nenes, 2010; Costantino et al., 2010; 2013~~ (Twomey, 1977; Feingold, et al., 2001~~). Another component of RFaci are rapid adjustments which may also lead to the modification of other cloud properties in response to the increase of N_d and decrease in CER, such as a ~~The second indirect effect describes the~~ decrease in precipitation efficiency~~ due to the decrease in the size of the cloud droplets~~, resulting in the increase of the LWP and the amount of clouds, thus enhancing the reflection of solar radiation (Albrecht, 1989). These two ~~first and second indirect~~ effects of aci are also referred to as the cloud albedo and cloud lifetime effects (Quaas et al., 2008).

The CER is an important factor affecting cloud physical processes and optical properties~~, which in turn influence precipitation and the Earth' radiation balance~~. Slingo (1990) pointed out that a reduction in the average CER by 15% - 20% can balance the radiative forcing at the top of the atmosphere caused by a doubling of carbon dioxide. Therefore, small changes in cloud microphysical properties may lead to important climate impacts (Zhao et al., 2018). Further study on the sensitivity of CER to ~~relations~~

~~between~~ aerosols ($S_{CER\text{-}A}$, further referred to as S) ~~and CER~~, together with meteorological parameters influencing ~~aerosol-cloud interaction~~aci, can improve our understanding of these processes and the effects of aci on RF, leading to improved aerosol-cloud parameterizations in regional climate models. The variation in N_d with CCN is referred to as the susceptibility β (β= $d \ln N_d / d \ln A$; e.g., Gryspeerdt et al. (2023)) and the variation of CER with CCN is referred to as the sensitivity S (eq. 1 in Section 3.1). Much of the variation of aerosol-cloud effective radiative forcing in ensembles of climate models is due to the variation in β, while β is also central to the strength of cloud adjustments (Gryspeerdt et al., 2023). The sensitivity of ~~effects of aerosols on the~~ microphysical ~~characteristics~~ properties of clouds to aerosol have been studied based on data from a large number of monitoring campaigns, using satellite, aircraft and ground based observations, and by using model simulations. Because of the large spatial coverage ~~and high spatial and temporal resolution~~, satellite instruments have been widely used to study aerosol-cloud interaction in different conditions, confirming the ~~large influence~~high sensitivity of ~~of aerosol particles on~~ cloud properties to aerosol (e.g., Yuan et al., 2008; Rosenfeld et al., 2014; Saponaro et al., 2017; Liu et al., 2018; Pandey et al., 2020; Christensen et al., 2020; Liu et al., 2021). In ~~In~~ studies on S ~~of the first indirect effect of aerosols with~~utilizing satellite data, which is the subject of the current study, the aerosol optical depth (AOD) is often used as a proxy for the aerosol concentration, which is justified by the correlation of AOD and CCN published by Andreae (2009). However, AOD is determined by all aerosol particles in the atmospheric column, including particles that do not act as CCN, depends on the relative humidity (RH) throughout the atmospheric column, does not provide information on chemical composition and may be influenced by aerosol in disconnected layers. The use of the Aerosol Index (AI), the product of AOD and the Ångström Exponent (AE; describing the spectral variation of AOD), is suggested as a better indicator of CCN because AE includes information on aerosol size (e.g., Nakajima et al., 2001). However, the AE is determined from AOD retrieved at two or more wavelengths and the evaluation of the results versus ground-based reference data shows the large uncertainty in AE. Therefore, in recent MODIS product Collections, AE is not provided over land (e.g., Levy et al., 2013; Kourtidis et al., 2015). AE is also not well-defined for low AOD for which uncertainty is largest (Bellouin et al., 2020; Gryspeerdt et al., 2023). The issues associated with using AOD or AI as proxy for CCN were discussed by, among others, Rosenfeld et al. (2014) who ~~-~~do not recommend the use of AI while also concluding that no better proxy is available. Therefore, in this study, AOD is used as a proxy for CCN to study S. It is noted that in other studies, e.g., Jia et al., 2022, both AOD and AI have been used and the results show similar behaviour.

Many ~~of these~~ studies confirmed the Twomey effect (e.g., Chen et al., 2014; Christensen et al., 2016; Jia et al., 2019). However, other studies show that, over some areas and especially over land in situations with high AOD, the CER increases with the increase of AOD, in contrast to the hypothesis of the ~~"~~"Twomey effect~~"~~" (e.g., Feingold et al., 2001; Yuan et al., 2008; Grandey and Stier, 2010; Tang et al., 2014; Wang et al., 2015; Jia et al., 2019; Liu et al., 2020). It is noted that in these studies, the relationship between CER and aerosol concentration was not constrained by LWP, although this is the premise of the Twomey effect~~first indirect effect of aerosol~~.

Meteorological conditions are important factors determining both the occurrence of clouds and cloud properties and therefore, in ~~aerosol-cloud interaction (ACI)~~aci~~ACI~~aci studies, the variation of meteorological conditions needs to be considered together with the variation of AOD (e.g., Myhre et al., 2007; Tang et al., 2014). On the one hand, ~~the~~ meteorological parameters ~~have the impacts on~~influence the Twomey effect. Jones et al. (2009) concluded that vertical motion, aerosol type, and aerosol layer height~~s~~ do make a significant contribution to RF_{aci}~~first aerosol indirect effect (AIE)~~ and that these factors are often more important than total aerosol concentration alone and that the relative importance of each differs significantly from region to region. ~~Su et al. (2010) studied~~ demonstrated ~~the influence of pressure vertical velocity (PVV) (ω_{700hPa}) on the~~ S ~~first indirect effect of aerosols and demonstrated the effect of this parameter on the CER and the LWP.~~ Wang et al. (2014) proved that the well-recognized aerosol effect mingles~~d~~ with meteorological conditions (RH and PVV), which likely is the main reason for the positive values of $S\beta_{\ln CER - \ln AOD}$ ~~(the change of CER with the change of AOD, see Section 3.1)~~ over land. Tang et al. (2014) observed the Twomey effect over ocean, but a positive CER-AOD relationship over Eastern China which they attributed to changes in relative humidity and wind fields. Tang et al. (2014) concluded that "o~~O~~ur results suggest that the effect of meteorology may not be negligible when investigating the aerosol indirect effect on a large scale, especially when the weather conditions are complex and change frequently.". Andersen and Cermak (2015) studied biomass burning aerosol over the Atlantic Ocean (Sep-Dec) in stable and unstable environments (LTS) and observed that the aerosol effect is stronger in unstable environment, especially during biomass burning episodes. These authors concluded that "the observed absolute differences in CER between stable and unstable environments are driven by cloud dynamical effects (CER and LWP are positively associated), or meteorology”. Jia et al. (2020) inferred that S increases remarkably with both cloud-base height and cloud geometric thickness (proxies for vertical velocity at cloud base), suggesting that stronger ~~aerosol-cloud interactions~~aci generally occurs under larger updraft velocity conditions. On the other hand, the meteorological parameters also ~~have the impacts on~~influence the potential adjustments. Koren et al. (2010) reported that observed cloud top height and cloud fraction correlate best with model pressure updraft velocity and relative humidity. Quaas et al. (2010) discussed the relationship between total cloud cover and AOD, often observed in satellite data, based on model simulations to test six hypotheses. These authors concluded that the increase of aerosol optical depth that accompanies the swelling of aerosol particles in humid airmasses is the dominant process contributi~~o~~ng to the observed correlation, confirming earlier conclusions by Myhre et al. (2007). Boucher and Quaas (2012) reported that aerosol humidification has a large impact on the relationship between AOD and rain rate and that discriminating the data into classes of pressure vertical velocity and/or relative humidity does not eliminate these meteorological effects. Gryspeerdt et al. (2014) studied the relationship between aerosol and initial cloud cover as a function of ~~relative humidity~~RH ~~(RH_{850hPa})~~ and vertical convection strength ~~(ω_{500hPa})~~. ~~Wang et al. (2014) proved that the well-recognized aerosol effect mingled with meteorological conditions (RH_{750hPa} and ω_{750hPa}), which likely is the main reason for the positive aerosol cloud interaction (ACI) index (defined in Section 3.1)values of $\beta_{\ln CER - \ln AOD}$ (the change of CER with the change of AOD, see Section 3.1) over land. Wang et al. (2015) discussed~~ S_{CER_A}~~the increase of CER with AOD in high AOD conditions over eastern China, which was observed during the summer but not in the winter, in terms of meteorological conditions. In particular they considered the different humidity effects during these seasons.~~ Liu et al. (2017) showed that ~~the formation of large cloud droplets in both horizontal and vertical directions and~~ the increase in cloud cover ~~are~~ is promoted in an environment with high ~~relative humidity~~RH ~~(RH_{950hPa})~~. A rising air mass ~~(ω_{750hPa})~~ can promote the formation of thicker and higher clouds. ~~Tang et al. (2014) observed the Twomey effect over ocean, but a positive CER-AOD relationship over Eastern China which they attributed to changes in relative humidity and wind fields. Tang et al. conclude that “Our results suggest that the effect of meteorology may not be negligible when investigating the aerosol indirect effect on a large scale, especially when the weather conditions are complex and change frequently.” Quaas et al.(2010), discuss the relationship between total cloud cover and AOD, often observed in satellite data,~~

~~based on model simulations to test six hypotheses. These authors conclude that the increase of aerosol optical depth that accompanies the swelling of aerosol particles in humid airmasses is the dominant process contribution to the observed correlation, confirming earlier conclusions by Myhre et al (2007. Boucher and Quaas (2012) report that aerosol humidification has a large impact on the relationship between AOD and rain rate and that discriminating the data into classes of pressure vertical velocity and/or relative humidity does not eliminate these meteorological effects. Andersen and Cermak (2015) studied biomass burning aerosol over the Atlantic (Sep-Dec) in stable and unstable environments (LTS) and observed that the aerosol effect is stronger in unstable environment, especially during biomass burning episodes. These authors concluded that "the observed absolute differences in CER between stable and unstable environments are driven by cloud dynamical effects (CER and LWP are positively associated, or meteorology".~~

The above are examples of ~~Only few~~ studies ~~have~~ address~~ed~~ing the influence of different aerosol and meteorological parameters on the sensitivity of cloud parameters to aerosol and potential confounding effects. M~~indirect effect and m~~ost of them used traditional statistical methods or stratified the data according to confounding meteorological parameters (~~-~~e.g., Saponaro et al., 2017; Ma et al., 2018). In the current study the geographical detector method (GDM) is applied as a complementary tool to quantify the relative importance of the effects of nine parameters on ~~ACI~~S. The GDM is explained in detail in Section 3.2. In brief, ~~is~~ a set of statistical methods is used to detect the spatial variability of aerosol and cloud properties, which are spatially differentiated, and evaluate the occurrence of correlations in their behaviour and the driving forces behind these correlations (Wang and Hu, 2012; Wang et al., 2016). The basic idea of the GDM is that the spatial distributions of two variables tend to be similar if these two variables are connected (Zhang and Zhao, 2018). The method ~~can be~~is used in this study to analyse the relative importance of different ~~influencing~~ factors, and interactions between them, influencing ~~on~~ ~~ACI~~aci.

The focus of the current study is to establish a CER-aerosol parameterization scheme by the application of the GDM to satellite data over two contrasting areas, i.e. the Yangtze River Delta (YRD) in eastern China, with high aerosol concentrations, and a relatively clean area over the East China Sea (ECS). The satellite data are first used to study the ~~build a relationship between~~ CER sensitivity to ~~and the~~ aerosol ~~concentration (using AOD as a proxy)~~ for different AOD regimes and all LWP values, followed by constraining the LWP in different intervals. It is noted that RF_{aci} is formulated in terms of N_d ~~the cloud droplet number concentrations N_d~~, whereas studies on the Twomey effects often use CER instead of N_d. CER is readily available as a satellite retrieval product, although in particular over land the reliability is questioned (Grandey and Stier, 2010), whereas N_d is derived from CER and the cloud optical thickness (COT) (e.g., Grandey and Stier, 2010; Arola et al., 2022). This implies that N_d is subject to the same retrieval errors as CER, including a possible relation between CER and LWP. The comparison of global maps of the sensitivities of CER and N_d to AOD by Grandey and Stier (2010) exhibits very similar patterns. In this study, the CER sensitivity to AOD is stratified by LWP, which however poses problems in the evaluation of RF_{aci}. However, the current study focuses on understanding effects of different parameters on CER sensitivity to aerosol rather than the application to determine RF_{aci}.

The results from the CER sensitivity study are used to guide ~~Next~~ the application of GDM ~~is applied~~ to determine the relative effects of different parameters on ~~the ACI~~aci. Relations between CER and AOD, meteorological conditions and several cloud properties are ~~analysed~~determined, including combined effects of different influencing parameters.

2 Approach

2.1 Study area

The complex aerosol composition and the high aerosol concentrations render eastern China an interesting area for a variety of studies of processes involving aerosols, including the current study on the use of satellite data for the systematic assessment of ~~aerosol indirect effects~~aci, i.e., S, adjustments and confounding meteorological factors~~.~~. The ~~ACI~~ study focuses on two areas, i.e. the Yangtze River Delta (YRD, ~~27~~26°N-35°N; ~~114~~113°E-122°E) in eastern China and the East China Sea (ECS, ~~20~~19°N-28°N; ~~126~~125°E-134°E). The locations of the YRD and the ECS are shown in the map in Figure 1.

The YRD has a developed economy, with much industrial activity, large harbors (sea and river) and related busy ship traffic, dense population~~s~~ in large urban centers, all with high traffic intensity and high energy consumption. In addition to the direct emission of black carbon, also aerosol precursor gases such as NO_2, SO_2 and VOCs are emitted from the combustion of biomass, coal and petrochemical fuels, leading to the formation of secondary aerosol particles such as nitrate and sulfate aerosols, while agricultural activities result in the emission of dust, ammonia and biological VOCs (BVOCs) into the atmosphere. These activities and associated emissions result in the occurrence of high AOD over the YRD. Over the East China Sea (ECS) the main aerosol types are sea spray aerosol generated by the interaction between wind and waves and anthropogenic pollutants transported from the ~~land~~ Asian continent over the ocean in the East Asian outflow. During transport over hundreds of km, aerosol particles are removed by several processes such as dry and wet deposition and hence the aerosol concentrations decrease and the AOD becomes relatively low and is dominated by sea spray aerosol. In view of the differences in aerosol composition and concentrations, the polluted YRD area and the relatively clean ECS area were selected as contrasting regions for the study of the influence of aerosols on cloud properties over land and over ocean.

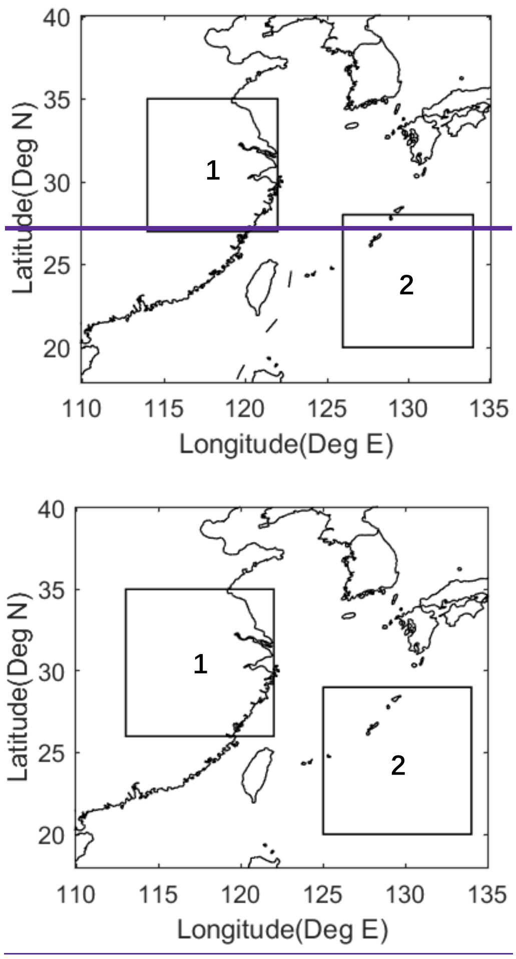


Figure 1. Map showing the locations of the two study areas selected for aerosol - cloud interaction studies: area 1 is the Yangtze River Delta (YRD; ~~27~~26°N-35°N, ~~114~~113°E-122°E), and area 2 indicates the selected Eastern China Sea area (ECS~~;) area (~~~~20~~19°N-28°N, ~~126~~125°E-134°E).

2.2 Data used

In this study, aerosol and cloud properties were used which were derived from measurements from the Moderate Resolution Imaging Spectroradiometer (MODIS) on-board the Aqua satellite, for the period 2008-2022 (15 years). This data was selected because the MODIS data are widely used and therefore they are well-characterized. In addition, the Aqua satellite flies in an afternoon orbit with local overpass time around 13:30, when the atmospheric boundary layer is well-developed. MODIS L3 C~~c~~ollection 6.1 daily aerosol and cloud parameters were downloaded from the LAADS website (Liu, 2022a~~https://ladsweb.modaps.eosdis.nasa.gov/, last access: 12 July 2022~~) with a spatial resolution of 1°x1°. Aerosol retrieval is only executed in clear sky conditions whereas cloud properties can only be retrieved in cloudy skies. Hence, it is not possible to– obtain co-located aerosol and cloud data from satellite. For satellite-based aci studies it is assumed that, following, e.g., Jia et al. (2022), aerosol properties are homogeneous enough to be representative for those in adjacent cloud areas. Consequences of this assumption were discussed by McComiskey and Feingold (2012). ~~The utilization of the daily MODIS aerosol and cloud data at 1°x1° resolution ensures that they are coincident when investigating aerosol - cloud interaction.~~ The MODIS instrument has 36 spectral bands - aerosol properties are retrieved using the first seven of these (0.47-2.13 μm) (Remer et al., 2005; Levy et al., 2013; Sayer et al., 2014; 2017) while additional wavelengths in other parts of the spectrum are used for the retrieval of cloud properties (Platnick et al., 2003; 2017). ~~More d~~Detailed information on algorithms for the retrieval of aerosol and cloud properties is provided at –http://modis-atmos.gsfc.nasa.gov (last access: 01 July 2021). In this study we use the AOD at 550 nm (referred to as AOD throughout this manuscript), CER, ~~cloud optical thickness (~~COT~~)~~, cloud liquid water path (LWP), cloud top pressure (CTP), cloud fraction (CF) and cloud top temperature (CTT).– The MODIS Collection 6.1 AOD product over China has been validated by, e.g., Che et al. (2019) and globally over land and ocean by Wei et al. (2019). MODIS C6.1 cloud products were evaluated by Platnick et al. (2017). The validation of CER and LWP, the primary cloud products used in this paper, was described by Painemal and Zuidema (2011), who compared MODIS C5 with in situ data (aircraft), and likewise the MODIS C6.1 CER product was evaluated by Fu et al. (2022) by comparison with airborne measurements. Fu et al. (2022) concluded that their "validation, along with in situ validation of MODIS CER from other regions (e.g., Painemal and Zuidema, 2011; Ahn et al., 2018), provides additional confidence in the global distribution of bias-adjusted MODIS CER reported in Fu et al. (2019)." It is noted that COT and CER are retrieved whereas LWP is secondarily derived (e.g., Painemal and Zuidema, 2011). AOD is used as a proxy for the amount of ~~aerosol particles~~CCN in the atmospheric column to investigate ~~ACI~~ aci (Andreae, 2009~~; Kourtidis et al., 2015~~) which seems to be the best alternative (Rosenfeld et al., 2014). As discussed in the Introduction, the use of an AE-based correction is not recommended over land (e.g., Kourtidis, et al., 2015). Comparisons with surface-based sun photometer data ~~revealed~~shows that Collection 6 ~~should~~ improves upon Collection 5, and overall, 69.4% of MODIS Collection 6 AOD feall within ~~an~~the expected uncertainty of ± (0.05 + 15%) (Levy et al., 2013; Tan et al., 2017). To reduce a possible overestimation of the AOD (e.g., due to cloud contamination), cases with AOD greater than 1.5 were excluded from further analysis. The choice of this threshold, rather than 0.6 used by Brendan et al. (2006), who used MOD06 Collection 04 products, is based on reports by Christenson et al. (2017) and (Varnái and Marshak, 2009). Christenson et al. (2017) used MOD06 C6 data (1km x1km) and reported that "large aerosol optical depths remain in the MODIS-observed pixels near cloud edges, due primarily to 3-D effects (Varnái and Marshak, 2009) and the swelling of aerosols by higher relative humidity." Varnái and Marshak (2009) noted that beyond 15 km contamination effects were minimized in MODIS data (1km x1km). Furthermore, we discarded scenes (1° by 1°) in which the aerosol distribution is heterogeneous, i.e. with a standard deviation higher than the mean value (Saponaro et al., 2017; Jia et al., 2022). As most aerosol particles are located in the lower troposphere (Michibata et al., 2014), the focus in this study is on warm clouds with CTT larger than 273K, CTP larger than 700 hPa and ~~LWP smaller than 200 g m^{-2}.~~LWP larger than 200 g m^{-2} is excluded to avoid deep convective clouds (Wang et al., 2014). Transparent-cloudy pixels (COT<5) were discarded to limit uncertainties (Grosvenor et al., 2018). The solar zenith angle was restricted to SZA < 65° and the viewing zenith angle to VZA <55° to avoid the large biases in COT and CER retrievals at larger angles (Grosvenor et al., 2018). ~~Cloud parameters were only considered in single liquid layer clouds.~~To ensure that the data used only included ~~consisted of~~ single layer liquid ~~layer~~ clouds and ~~, as well as~~ nonprecipitating cases, ~~we applied~~ the ~~same~~ filtering criteria ~~as~~described by Saponaro et al. (2017) were applied~~to the MODIS cloud data.~~ ~~Yuqin works on precipitation filtering: reprocess)~~

Confounding ~~Effects of~~ meteorological ~~conditions~~ effects on ~~ACI~~ aci were explored using the daily temperature at the 700 and 1000 hPa levels, ~~relative humidity (~~RH~~)~~ at the 750 hPa level and ~~pressure vertical velocity (~~PVV~~)~~ at the 750 hPa level. Low tropospheric stability (LTS), which is defined as the difference in potential temperature between the free troposphere (700 hPa) and the surface (1000 hPa), is used as a measure of the strength of the inversion that caps the planetary boundary layer (Klein and Hartmann, 1993; Wood and Bretherton, 2006). These meteorological data were retrieved from the ECMWF ERA-5 reanalysis data which provide global meteorological conditions at 0.25°x0.25° resolution for 37 pressure levels in the vertical (1000-1 hPa), for every 1 h (UTC) (Liu, 2022b~~https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels-monthly-means?tab=form, last access: 12 July 2022~~). The meteorological parameters were resampled to the MODIS/Aqua overpass time at 13:30 (local time) by taking a weighted average ~~of the properties~~ at the two closest times (05:00 UTC and 06:00 UTC) provided by the ECMWF ERA-5 reanalysis data~~ERA Interim~~.

Table 1. ~~The list of the p~~Parameters used ~~, sources, and their corresponding temporal-spatial resolutions applied~~ in the present study, together with the sources, time periods and spatial resolutions.

Source	Time period	Resolution	Parameters
MYD08	Jan 2008-Dec 2022	Daily, 1°x1°	AOD at 550 nm
			COT at 2.1 um
			CER at 3.7 um and 2.1 um
			Cloud-top temperature
			Cloud-top pressure
			LWP at 2.1 um
			Cloud Fraction
			Solar zenith angle
			Sensor zenith angle
			Cloud multi-layer flag
			Cloud phase flag
ERA5	Jan 2008-Dec 2022	hourly, 0.25°x0.25°	Temperatures at 700 and 1000 hPa
			Relative humidity at 750 hPa
			Vertical velocity at 750 hPa

3 Methods

3.1 Sensitivity of ~~Aerosol-~~cloud ~~interaction index~~parameters to changes in aerosol concentrations

~~The first indirect effect of aerosols is defined as the variation of c~~Changes in aerosol loading lead to an adjustment of cloud optical or microphysical parameters (~~cloud optical thickness, cloud droplet effective~~

~~radius~~COT, CER, etc.)~~ with the variation of aerosol loading~~. Aerosol particles can become ~~cloud condensation nuclei (~~CCN~~)~~ or ~~ice nucleating particlesi (~~INP~~)~~, depending on their chemical composition and ambient temperature. When these nuclei are activated, they become cloud droplets due to condensation of water vapor. When the concentration of aerosol particles increases, often also the number of CCN or INP may increase~~s~~ and thus the number of cloud droplets may increase. However, if the liquid water content in the cloud does not change (as indicated by a constant LWP), the condensable water will be distributed over more cloud droplets which thus remain smaller, i.e. the CER decreases and the cloud albedo increases when the aerosol concentration increases. On the basis of findings of Kaufman and Fraser (1997), Feingold et al. (2001) pointed out that the sensitivity of cloud microphysical properties (e.g. CER) to changes in aerosol (e.g., AOD) can be described by ~~aerosol-cloud interaction caused by the first indirect effect can be calculated by~~ the following formula:~~:~~

$$S=S_{CER\text{-}A}\cancel{IE=ACI_r}=-\frac{d\ln r_e}{d\ln\alpha}\Big|_{LWP} \quad 0<\cancel{SACI_r}<-0.33 \quad (1)$$

Where r_e represents the ~~cloud droplet effective radius (~~CER~~)~~ and α represents the ~~aerosol number concentration~~AOD. Following Andreae (2009), AOD and CCN are correlated and AOD varies with CCN following a power law relationship. ~~Equation~~ Eq. (1) describes the relative change of CER with the relative change of the ~~aerosol concentration~~AOD for constant LWP. It is noted that this formulation differs from that used in recent studies (e.g., Bellouin et al., 2020) where S is expressed in ~~the cloud droplet number concentration~~ N_d with no restriction in LWP. The sensitivity ~~In this study, the AOD is used as a proxy for the aerosol number concentration and ACI$_r$~~ S of CER to AOD can be determined as the slope of a linear fit to a log-log plot of CER versus AOD. It is noted that S~~ACI$_r$~~ is a function of CER and effects on CER directly influence S~~ACI$_r$~~. In this study effects on ~~ACI$_r$ (from here on simply denoted as SACI)~~ and CER are used interchangeably~~intermittently~~. Relations between CER and AOD are determined through Eq. 1 and correlation coefficients R. The significance of these ~~co~~rrelations i~~was~~ determined by using the student's t test, i.e. the results are statistically significant when the p value is smaller than 0.01, where p is defined as the probability of obtaining a result equal to or "more extreme" than what was actually observed.

3.2 Geographical detector method

The geographical detector method (GDM) is introduced to analyze which factors influence the ~~ACI~~aci and identify possible correlations between different factors. The GDM is based on the assumption that if an independent variable has an important influence on its dependent counterpart, their spatial distributions should also have evident similarities (Wang and Hu, 2012; Wang et al., 2016). ~~It~~The GDM not only ~~takes~~ accounts ~~of~~for the rank order of the variables as determined by the Spearman's Rank method but also spatial information. The geographical detector provides four modules, including ~~the~~ factor detector, interaction detector, risk detector and ecological detector. In this study, the first two modules are used to detect interactions between different parameters, based on their spatial variations, and thus reveal the driving factors for aerosol-cloud interaction over the target regions. The influencing factors (x) considered in this study are aerosol and meteorological parameters and the dependent factors (y) are ~~the ACI index~~S and cloud parameters. In the GDM, for example, the CER data are recorded in a raster grid as illustrated in Figure 2. The data in the raster grid is transformed into dot files, each dot containing a value for the CER and for one of the influencing parameters x. The dependent (CER) and influencing (x) parameters are separated into 2 layers with the same grid. In the x~~x~~ layer, the Jenks natural breaks classification method (Brewer and Pickle, 2002), aiming to minimize the variance within the group and maximize the variance between groups, was applied to categorize the whole region into ~~N~~i sub-regions (3 in Figure 2), according to pre-defined ranges of influencing factors (e.g., AOD). In each sub-region~~spatial unit (N_i)~~, the influencing factor (x) varies within certain limits, with variance σ_i. The power of determination q of x~~x~~ to y~~y~~ (also referred to as power of the influencing factor) determines the extent to which a factor (x) influences the dependent factor (y) over the whole study area and is calculated using ~~Equation~~ Eq. (2):

$$q = 1 - \frac{\sum_i^L N_i \sigma_i^2}{N\sigma^2} \qquad (2)$$

where i (1, …, L) is the number of sub-regions of factor x~~x~~; N represents the total number of spatial units over the entire study area; N_i denotes the number of samples in sub-region i; and σ_i^2 and σ^2 denote the ~~s~~ variance of the samples in the subregion i and the total variance in the entire study area, respectively. The value of q~~q~~ varies between 0 and 1, i.e. q[0,1], where 0 indicates that factor x~~x~~ has no influence on y and the closer q is to 1, the greater the influence of x~~x~~. For instance, if $q = 0.5$, x~~x~~ can explain 50% of the variation of y~~y~~. In this study, multi-years of mean values of influencing factors (x) and dependent factors (y) we~~a~~re calculated for each raster grid. Then, we classified the influencing factors (e.g. AOD and meteorological parameters) into 5 sub-regions by the Jenks natural breaks classification method (Brewer and Pickle, 2002). For example, AOD needs to be classified into 5 levels using the Jenks natural breaks classification method, and the AOD source data needs to be reclassified into 1-5 natural numbers from small to large, and then counted into the grid. Therefore, the input of the independent variable AOD is a type variable. However, it should be noted that the GDM also has unstable characteristics. On the one hand, it is due to the MAUP (Modified Area Unit Problem) variable area unit problem, which can be understood as the influence of "scale effect". Due to the limitation of data resolution used in this study, the spatial statistical unit is 1°x1°. On the other hand, the methods used for data discretization can also have an impact. This study attempts to determine the optimal number of classifications by examining the impact of number of classification levels (3-8) on the GDM output results. The results show that the number of classification levels does not affect the relative importance of cloud factors on the cloud. Here we classify the values of each cloud factor into 5 levels during the period of 2008-2022.

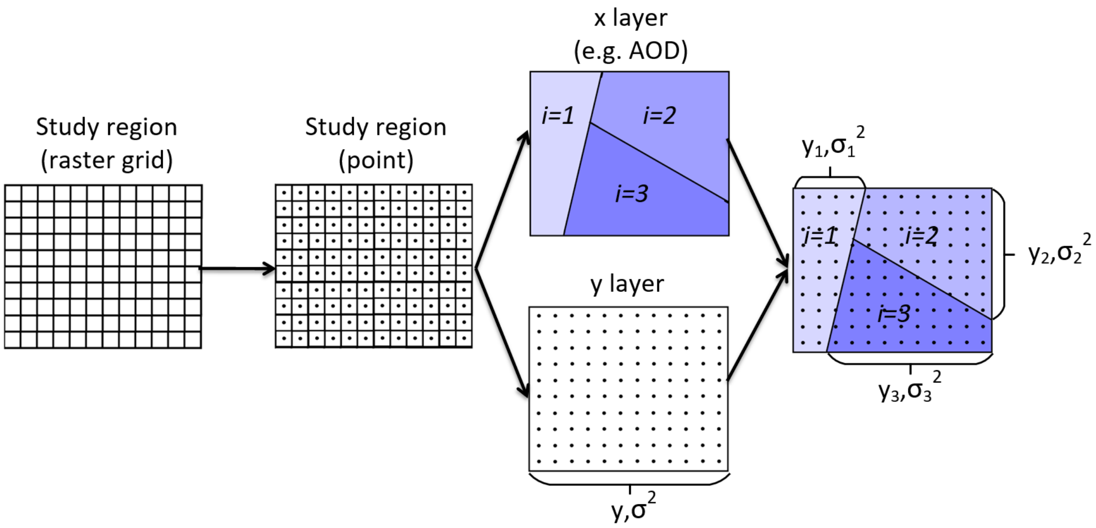


Figure 2. The principle of the geographical detector method. See text for explanation.

The interaction detector can be used to test for the influence of interaction ~~of~~between different influencing factors ~~and to determine whether the interaction of two factors~~, e.g., $x1$ and $x2$, on the dependent factor (y~~y~~) and whether this interaction weakens or enhances the influence of each of x~~x~~1 or x~~x~~2 on the dependent variable, y, or whether they are independent in influencing y. For example, Figure 3(a) shows~~ed~~ the spatial distribution of the dependent variable, y. The factors $x1$ and $x2$ both vary across the study region, but in different ways, and for each factor different sub-regions can be distinguished by application of the Jenks classification method described above to each factor separately. This is ~~as~~ illustrated in Figures 3(b) and 3(c) where, as an example, ~~for each factor~~ three different sub-regions are considered for each factor~~),~~ ~~as shown in Figure 3(b) and 3(c).~~ Usually, the dependent variable y~~y~~ is influenced by several different factors xi~~x_i~~ (Figure 3) and the combined effect of two or more factors may have a weaker or stronger influence on y than each of the individual factors. The q values for the influences of factors x~~x~~1 and x~~x~~2 on y~~y~~, obtained from the application of the factor detector method (~~equation~~ Eq. 2), may be represented as q (x~~x~~1) and q (x~~x~~2). Hence, a new spatial unit and subregions may be generated by overlaying the factor strata x~~x~~1 and x~~x~~2, written as x~~x~~1∩x~~x~~2, where ∩ denotes the interaction between factor strata x~~x~~1 and x~~x~~2 as illustrated in Figure 3(d). Thus, the q value of the interaction of x~~x~~1∩x~~x~~2 may be obtained, represented as q (x~~x~~1∩x~~x~~2). Comparing the q~~q~~ value of the interaction of the pair of factors and the q~~q~~ value of each of the two individual factors, five categories of the interaction factor relationship ~~are described~~can be considered which are ~~as~~ summarized in Table ~~1~~2. If q~~q~~(x~~x~~1∩x~~x~~2) > q~~q~~(x~~x~~1) + q~~q~~(x~~x~~2), this is referred to as a nonlinear enhancement of two variables. And if $q(x1∩x2)$ > Max[$q(x1)$, $q(x2)$], this is referred to as a bilinear enhancement of two variables. The occurrence of nonlinear enhancement and bilinear enhancement ~~is~~are indicated with the q~~q~~ values in Table 2 and in the caption of Figure 7~~in the revised manuscript~~.

It is noted that the q~~q~~-values of multiple influencing factors are considered separately they may sum up to larger than 100%. However, when the variables are correlated they must be considered together and the interaction q~~q~~-value must be evaluated.

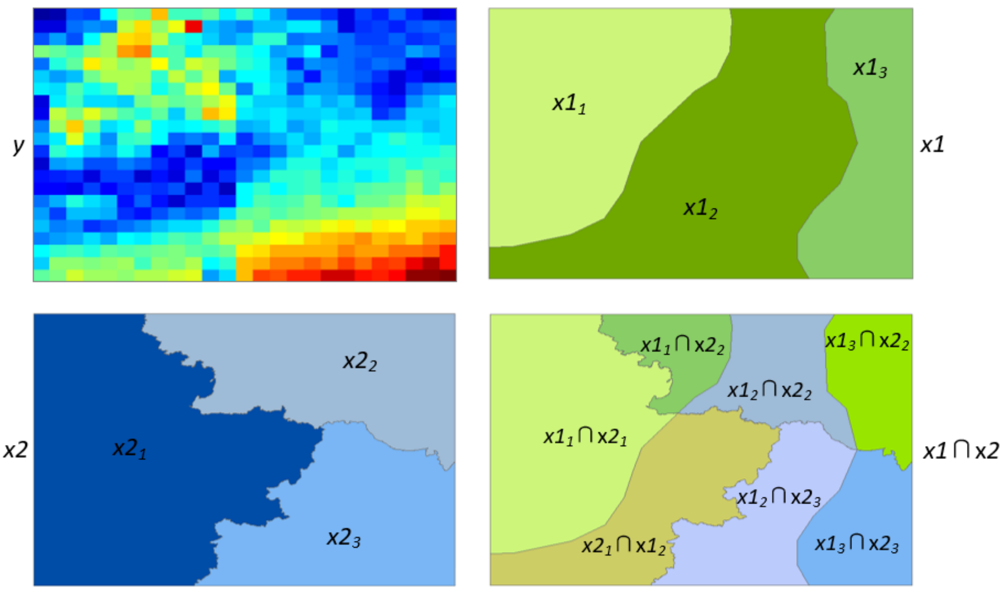


Figure 3. Detection of interaction (see text for explanation).

Table ~~1~~2. Interaction categories of two factors and the interaction relationship

Illustration	Description	Interaction
	$q(x1 \cap x2) < Min[q(x1), q(x2)]$	Weakened, nonlinear
	$Min[q(x1), q(x2)] < q(x1 \cap x2) < Max[q(x1), q(x2)]$	Weakened, unique
	$q(x1 \cap x2) > Max[q(x1), q(x2)]$	Enhanced, bilinear
	$q(x1 \cap x2) = q(x1)+ q(x2)$	Independent
	$q(x1 \cap x2) > q(x1)+ q(x2)$	Enhanced, nonlinear

The geographical detector method has been used to detect influencing factors for several different purposes (e.g., Wang et al., 2018; Zhang and Zhao, 2018; Zhou et al., 2018). For example, the GDM ~~A geographical detector~~ was used to detect the influence of ~~tial~~ annual and seasonal factors on the spatial-temporal characteristics of surface water quality (Wang et al., 2018). Other examples are the application of the GDM to examine ~~Moreover, the driving factors' influences~~ factors influencing~~on~~ regional energy-related carbon emissions (Zhang and Zhao, 2018) and to examine ~~the~~ effects of socioeconomic development on fine particulate matter (PM2.5) in China (Zhou et al., 2018)~~ were examined by the geographical detector technique.~~ In the current study, the GDM ~~is method~~ was used to detect the impact of nine variables and their interactions on ~~the variations of ACI~~S and cloud parameters over land and ocean. The advantages of using the GDM in this approach are (1) stratified independent variables enhance the representation of a sample unit, so it has higher statistical accuracy than other models with the same sample size; (2) the use of a q-statistic value can afford a higher level of explanatory power, but does not require the existence of a linear relationship between independent and dependent variables; (3) the ~~geographical detector~~GDM can determine the true interaction between two variables and is not limited to pre-established multiplicative interactions (Wang et al., 2010); (4) the use of ~~a geographical detector~~the GDM does not need to consider the collinearity of multiple independent variables (Wang et al., 2010).

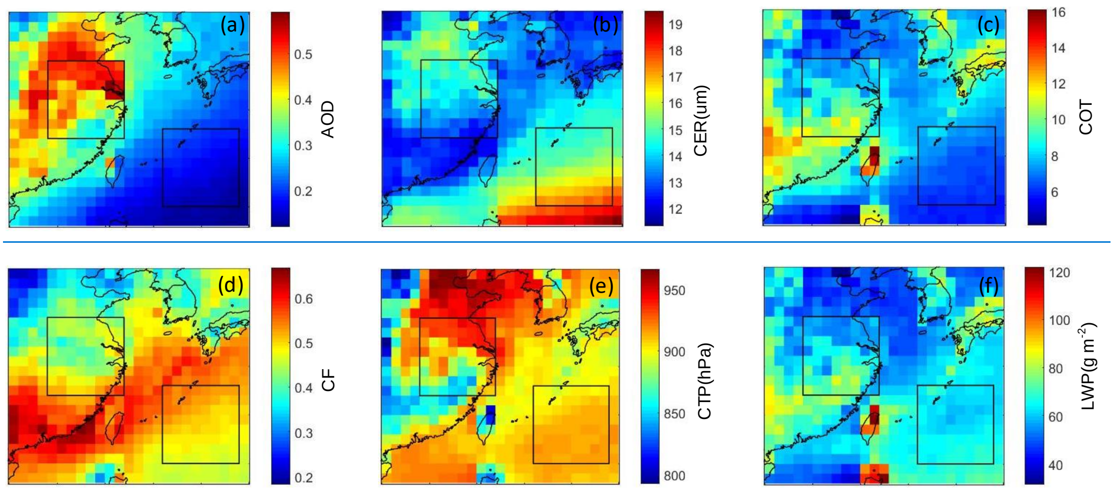
4 Results ~~and Discussion~~

4.1 Spatial distribution and correlation analysis of AOD and cloud parameters

The spatial variations of the AOD and the cloud properties (CER, COT, CF, CTP and LWP) over the study area, averaged over the years 2008-2022, are presented in Fig. 4. Figure 4(a) shows a large difference between the AOD over land and ocean, with the highest values over the northern part of the

YRD (averaged AOD larger than 0.5), and the lowest values over the southwestern part of the ECS (<0.1); the AOD decreases gradually from land to ocean. The spatial distributions of the CER, COT, CF, CTP and LWP~~)~~ over the YRD and ECS in Figs. 4(b)–(f) shows that for each of them there is a distinct difference between those over land and over ocean both as regards the values and the spatial variation. Over the ECS, the CER is largest in the south and decreases toward the north of this area and the values are overall substantially larger than over the YRD, where the CER varies somewhat and decreases from north to south. The variation of the CER with AOD over the YRD is opposite to what would be expected, which will be discussed in Sect. 4.2. The COT also varies somewhat over the YRD, but~~,~~ contrary to the CER, COT increases from north to south. Over the ECS, the COT is generally lower than over the YRD, with the highest values in the northwest~~NW~~ which gradually decrease toward the ~~SW~~southwest. Clearly, the CER is higher and the COT is lower over the ECS than over the YRD.

The spatial distributions of CF, CTP and LWP are clearly different. Over the ECS, CF increases from the ~~SW~~ southwest to the ~~NW~~northwest, opposite to the variations of the CTP and the LWP which are lower in the north of the ECS than in the south. Over ocean the clouds are generally lower (higher CTP) than over land, and CTP varies over the study area with the highest values over land, in the north. Over the YRD, the spatial patterns of the CF and CTP are opposite, with CF increasing from south to north and CTP decreasing. Over the YRD, the spatial distributions of COT and LWP are similar with higher values toward the south. Over the ECS, the LWP varies with the lowest values in the ~~NW~~ northwest and the highest values in the south. The high values of the CER and CF over the ECS could be due to the dominance of sea spray aerosol, the high hygroscopicity of which makes these particles very efficient CCN. The influence of different factors on the sensitivity of cloud parameters to aerosol and the adjustments are discussed in the following sections, based on both statistical methods and the application of the GDM.



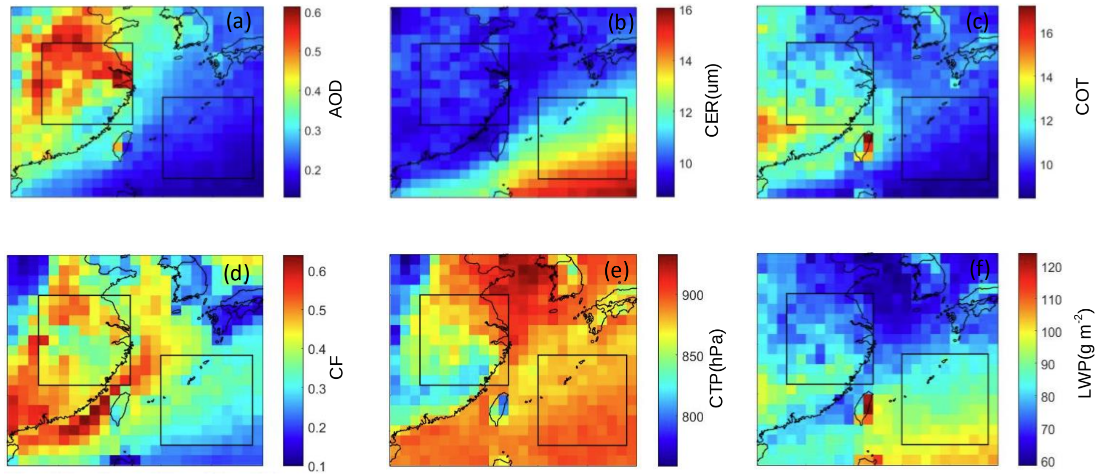


Figure 4. Spatial distributions of AOD (a), CER (b), COT (c), CF (d), CTP (e) and LWP (f), averaged over the years 2008 – 2022, over the study area, with the YRD and ECS marked by the squares.

4.2 Sensitivity of ~~ACI estimates for~~ CER to AOD ~~for the whole LWP regime~~

~~Equation~~ Eq. (1) shows that the value of ~~the ACI~~the sensitivity S of CER to AOD is determined by the slope of a linear fit to a log-log plot of CER versus AOD. To investigate S~~the ACI~~, we used correlated data pairs for 15 years and the data was binned in AOD intervals with a bin width of 0.02, and the CER data in each AOD bin were averaged. Logarithmic plots of the averaged CER data versus AOD over the YRD and the ECS are presented in Figure 5. Figures 5(a)~~ (YRD)~~ and 5(b)~~ (ECS)~~ show different regimes for the variation of the CER with the AOD over the YRD and the ECS. The first regime, for AOD ≤ 0.05, shows the increase of CER with AOD over both regions, followed by a variable CER over the YRD and a gradually stronger decrease over the ECS for AOD between 0.05 and 0.1. In view of this variability and the uncertainty of AOD of ± (0.05 + 15 %) over land and ± (0.03 + 5 %) over ocean (Levy et al., 2013), ~~the ACI~~S will not be investigated for AOD < 0.1. For higher AOD, S~~the CER / AOD relation~~ changes for AOD around 0.3. Thus, the second regime is selected as the part of the CER vs AOD relationship where~~for~~ AOD varies between 0.1 and 0.3. In this AOD regime, the CER fluctuates a little with AOD over the YRD (Figure 5(a)) ~~but the CER values remain within the standard deviation~~ and S is close to 0 (no ~~the expected~~discernible Twomey effect) ~~is not observed~~. In contrast, over the ECS the CER clearly decreases with AOD for AOD increasing from 0.1 to 0.3 (Figure 5(b)), in good agreement with expectation based on the Twomey effect, and the correlation between CER and AOD is high with R=0.99 and statistically significant. Note however, that no selection was made for LWP and the condition of constant LWP was not fulfilled. This will be further discussed in Section 4.3.

In the third regime, where AOD > 0.3, CER increases with increasing AOD over the YRD, with correlation coefficient R= 0.79. In contrast, over the ECS the CER does not significantly change with increasing AOD for AOD>0.3 (very small S). However, the large uncertainty in the bin-averaged CER in this AOD regime, increasing with increasing AOD, indicates a very variable ~~ACI~~ S between high-AOD events which on a statistical basis cannot be further analysed and likely depends on the type of aerosol present during each event and the meteorological conditions. The reason for the ~~positive relationship~~increase of ~~between~~ CER ~~and~~ with increasing AOD (S positive) over the YRD may be similar to that described by Feingold et al. (2001), i.e., in the presence of a large number of aerosol particles (CCN) competing for a limited amount of water vapor, only a subset of aerosol particles is activated. Once activated, these particles continue to grow faster, thus preventing water vapor from condensing onto smaller aerosol particles that are less susceptible to activation. As a result, the amount of available water vapor is distributed over a subset of aerosol particles which thus become cloud droplets with relatively large CER and the CER in turn increases with further increasing AOD (Liu et al., 2017). The CER sensitivity ~~response of CER~~ to AOD is stronger over the ECS (0.1<AOD<0.3) than over the YRD (AOD>0.3). It is anticipated that during the relatively low AOD over the ECS ~~during~~ in AOD regime 2 (0.1<AOD<0.3~~AOD 0.1-0.3~~) the aerosol number concentration is dominated by sea spray aerosol particles (de Leeuw et al., 2011) which are hygroscopic and thus provide good CCN, while over open ocean also the ~~relative humidity~~RH is generally high. Hence the available water vapor will be readily distributed over all CCN, resulting in the decrease of the CER and a strong correlation with AOD. Further, the AOD over open ocean does not reach high values in the absence of continental influence, even in very high wind speeds the AOD does not exceed 0.2 (Huang et al., 2010; Smirnov et al., 2012). Hence AOD higher than 0.2 over the ECS is influenced by long-range transport of aerosol produced over land with lower hygroscopicity, and thus lower susceptibility to act as CCN, which explains the breakdown of the Twomey effect over the ECS for elevated AOD. In fact, the data in Figure 5(b) show that the CER~~/~~-AOD relationship starts to flatten for AOD ~0.2 and is flat for AOD larger than ~0.3. Overall, Figure 5 shows that the Twomey effect is clear in the second AOD regime over the ECS and the anti-Twomey effect in the third AOD regime over the YRD. For this reason, the further analysis focuses on the aci~~ACI~~ over the ECS for AOD between 0.1 to 0.3, and over the YRD for AOD > 0.3.

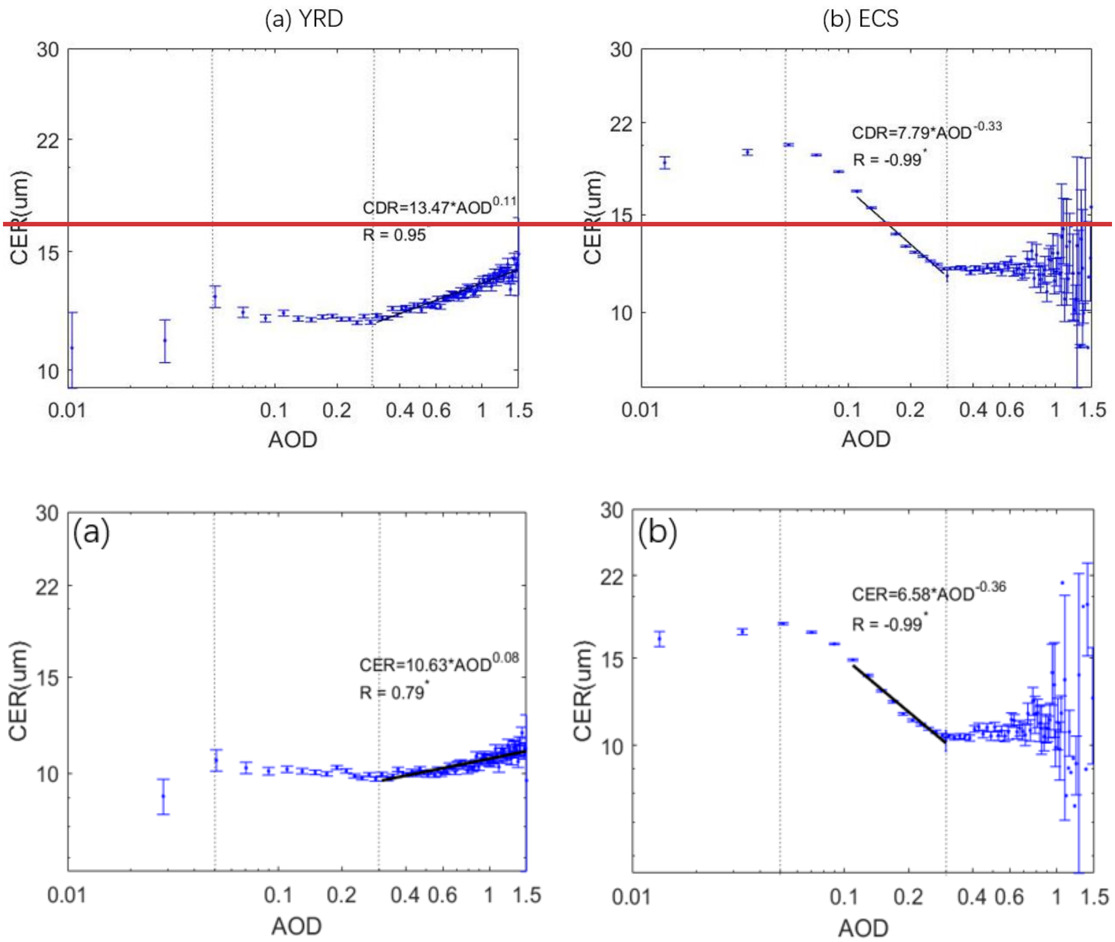
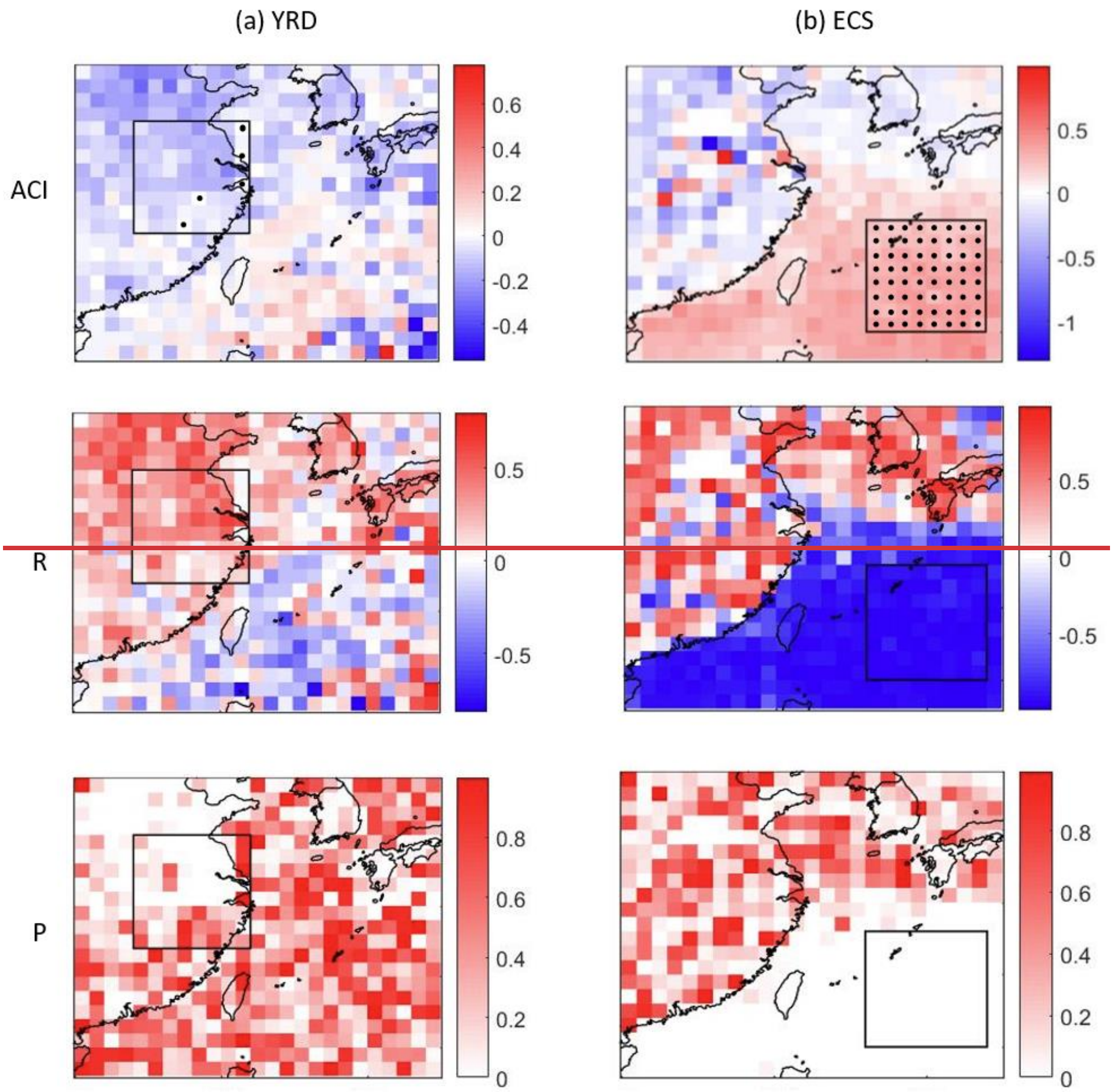


Figure 5. Variation of CER with AOD over the YRD (a) and the ECS (b). Here all CER data were averaged in AOD bins, from 0.0 to 1.5 with a step of 0.02. Note that the data are plotted on a log-log scale. The lines for the YRD data for AOD>0.3 and for the ECS data for 0.1<AOD<0.3 represent least-square fits to the binned data, and the resulting relations are presented in each figure. The marker ∗ at the top right corner of the R value indicates that the correlation is statistically significant with p < 0.~~05~~01. The thin vertical lines indicate the AOD regimes as explained in the text.

To study the spatial variation of ~~the ACI~~S over the study area, ~~the ACI~~S ~~in each grid cell~~ has been calculated in each grid cell by application of ~~Equation~~ Eq. (1) to all observations over the YRD for which AOD>0.3 and to all observations over the ECS for which $0.1<\mathrm{AOD}<0.3$~~)~~. The results are plotted in Figure 6, which shows maps of ~~the ACI~~S, the correlation coefficient R between CER and AOD and the statistical P-value for each grid cell over the study area. Figure 6(b) shows that ~~the ACI~~ over the ECS, for the second AOD regime (0.1-0.3), S is ~~positive~~negative, with large negative correlation coefficients (-0.~~78~~66~~–~~ to -0.~~99~~98) which mostly are statistically significant ($p < 0.01$). These results show the good correlation between CER and AOD, consistent with the cloud albedo effect~~theory of the first AIE~~. In contrast, over the YRD, for the third AOD regime (>0.3), ~~the ACI~~S is mostly ~~negative~~ positive and the correlation between CER and AOD is positive, i.e. high aerosol loading results in larger CER for AOD>0.3, as was also concluded from Figures 5(a) and (b). The data in Figure 6(a) also show that, ~~the negative~~ over the YRD, S is largest over the ~~CER/AOD relation is strongest in the part of the selected YRD region~~area to the north of Shanghai but R is relatively ~~small~~ weak (0.11 ~~–~~to 0.~~63~~35) and for the majority of the cells the correlations are not statistically significan~~ce is~~t (p ~ 0.1 or larger). ~~In the part of the selected YRD region s~~South of Shanghai the correlations are small and not statistically significant. The observed anti-Twomey effect of aerosols over the YRD has also been reported in earlier publications such as Jin and Shepherd (2008), Yuan et al. (2008) and Liu et al. (2017). Factors influencing the relationship between AOD and cloud parameters have been reported in the literature, such as hygroscopic effects (e.g., Qiu et al., 2017), atmospheric stability, cloud dynamics, cloud height (Shao and Liu, 2005) and land cover type (Jin and Shepherd, 2008; Ten Hoeve et al., 2011). The effects of competing mechanisms and their possible influence on the observed response of CER to high AOD in the YRD will be further discussed in the following sections.



(a) YRD
(b) ECS
ACI
R
P
0.6
0.4
0.2
0
-0.2
-0.4
0.5
0
-0.5
-1
0.5
0
-0.5
0.5
0
-0.5
0.8
0.6
0.4
0.2
0
0.8
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0.4
0.2
0

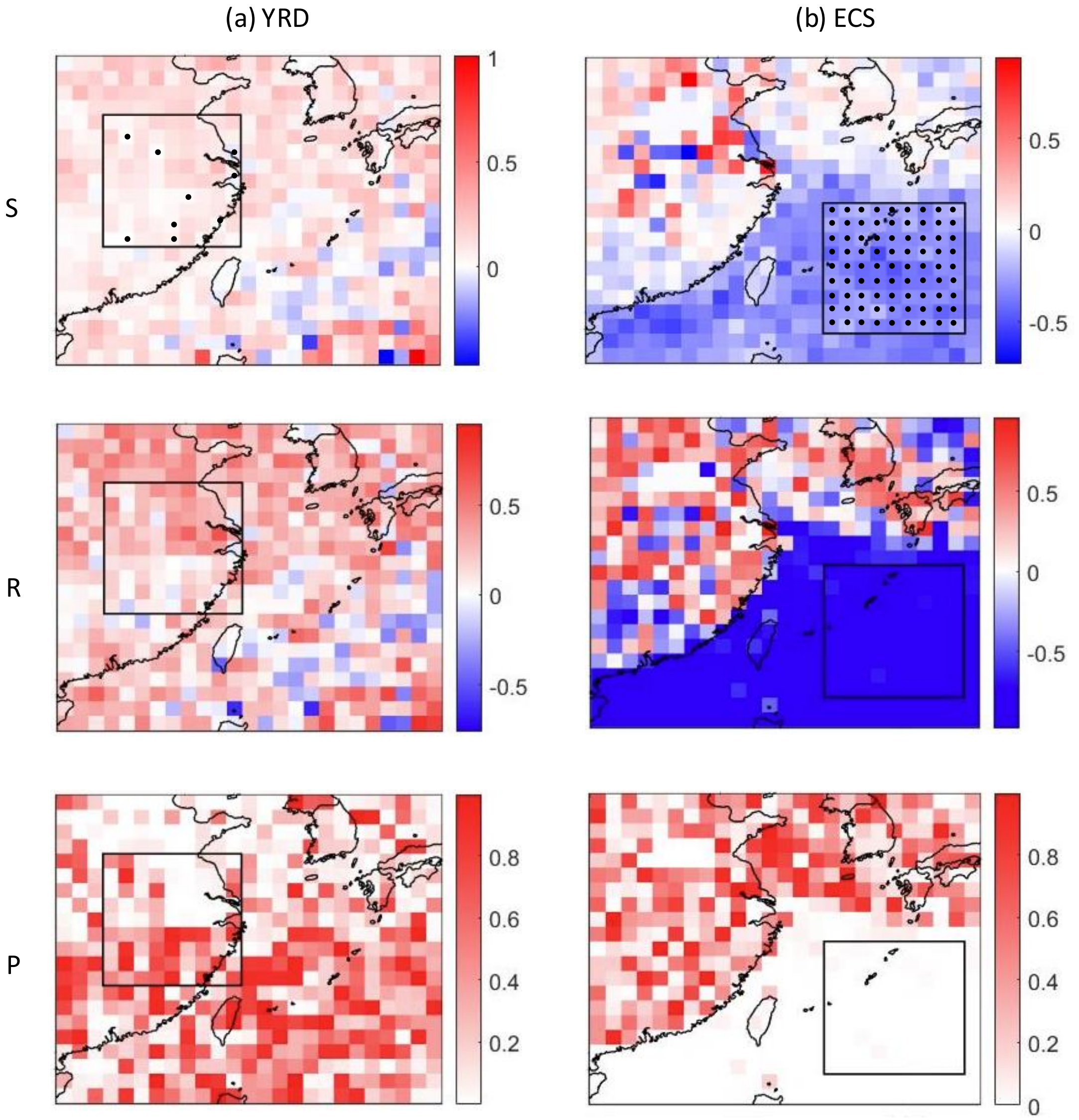


Figure 6. Using the AOD as a proxy for ~~aerosol concentrations~~CCN, estimates of the ~~ACI for~~ CER sensitivity to aerosol (S) were calculated for each grid point in both study areas. Maps of the spatial distributions of ~~the ACI~~S, the correlation coefficients and the statistical P-values in each grid point are presented ~~in Figure (a) for~~ over the YRD (left column, Figure 6(a)) for the AOD regime with AOD>0.3 and ~~in Figure (b)~~ over the ECS (right column, Figure 6(b)) for the AOD regime with 0.1<AOD<0.3 ~~(right column, Figure 6b)~~. ~~ACI~~S, R and P-values are color coded following the color bars at the right of each figure. The black solid dots in the top figures (S), indicate that the ~~ACI~~ S value is ~~positive~~negative in the grid point over the YRD and ECS.

4.3 Sensitivity of CER to AOD ~~ACI estimates for CER for different~~stratified by LWP ~~regimes~~

In the data presentation and discussion of S in Section 4.2, the condition of constant LWP ~~(Equation 1) was not considered. Because constant LWP is a condition~~ for the application of E~~e~~q. (1) and the occurrence of ~~to~~ the cloud albedo effect, was not considered.~~calculate ACI, in~~ In this Section ~~–~~the effect of LWP on S will be further investigated. To this end, the condition of constant LWP is approached by stratifying ~~considering~~ LWP into five ~~LWP~~ intervals, each with a width of 40 g m^{-2}, for the LWP ~~the~~ range of [0 g m^{-2}, 200 g m^{-2}]. ~~The ACI~~S ~~in the whole area~~ was calculated over the YRD and the ECS, for each LWP interval using Eq. ~~uation~~ (1) for all observations over the YRD for which $AOD>0.3$ and for all observations over the ECS for which $0.1<AOD<0.3$. The results are presented in Table ~~2~~3, together with the corresponding correlation coefficients R ~~for the relation~~ between ~~the~~ CER and AOD in the relevant AOD regimes. ~~The significance of these correlations was determined by using the student's t test, i.e. the results are statistically significant when the p value is smaller than 0.01, where p is defined as the probability of obtaining a result equal to or "more extreme" than what was actually observed.~~ The data in Table ~~2~~ 3 show that over the ECS, ~~the ACI estimates are~~S is negative ~~positive~~ and statistically significant for all four LWP ranges between 40 and 200 g.m^{-2}~~,~~. ~~and~~ The sensitivity becomes stronger as LWP~~increase with~~ increas~~ing LWP~~es, i.e., S changes from -0.19~~7~~ (LWP 40-80 g.m^{-2}) to -0.46~~2~~ in the highest LWP range (160-200 g.m^{-3}), with corresponding R of -0.98 to -0.99. Thus, the magnitude of the LWP has a substantial influence on the ~~aerosol~~ albedo effect~~ on the ACI~~. Over the YRD, ~~the ACI~~S is positive and statistically significant in the first three LWP regimes, with values ~~the ACI~~ varying between ~~-~~0.06~~10~~ and ~~-~~0.10~~3~~ and ~~, with~~ a correlation R between 0.57~~76~~ and 0.81~~96~~. These data show that, in contrast to the ECS, over the YRD the variation of the LWP ~~effect~~ has little influence on ~~the aci~~S~~ACI is smaller than over the ECS~~ and~~, in contrast to the ECS, over the YRD~~thus the magnitude of the LWP has little influence on the ~~strength of the ACI~~albedo effect.

In summary, the data show that both over the ECS and the YRD the relationships between the CER and the AOD are significant, but for different LWP intervals ([0 g m^{-2}, 120 g m^{-2}] over the YRD and [40 g m^{-2}, 200 g m^{-2}] over the ECS)~~)~~ and for different AOD regimes ($0.1<AOD<0.3$ over the ECS and $AOD>0.3$ over the YRD), and that the ~~aciACI~~CER-AOD relation follows the Twomey effect over the ECS and the anti-Twomey effect over the YRD.

The variation of S with changes in LWP indicates that the condition of constant LWP is not truly satisfied: if the data would be stratified according to smaller LWP intervals (quasi-constant LWP, Ma et al., 2018), S would likely vary more smoothly with LWP. As mentioned in the Introduction, LWP is not directly retrieved but calculated form CER and COT and thus also the calculation of S is to some extend affected by LWP. We further note the results by Ma et al. (2018), i.e. the slope of CER versus AI (comparable to S in this paper) varies little with LWP, with positive values over land and negative values over ocean and thus behaves similar to the data in Table 3 for YRD and ECS.

In the following study on the effects of the AOD and different cloud and meteorological properties on ~~the ACI~~S and adjustments~~, using the GDM~~, these differences will be taken into account, i.e. over the YRD only data with AOD > 0.3 and LWP in the range from 0 to 120 g m^{-2} will be used and over the ECS only data with AOD in the interval [0.1, 0.3] and LWP in the range from 40 to 200 g m^{-2} will be used.

Table ~~2~~3. ~~ACI e~~Estimates of S, computed using Eq. (1), and correlation coefficients R between~~or the~~ CER and ~~-~~AOD, ~~relationships in five different~~stratified by LWP, ~~LWP intervals computed using Equation (1)~~ over the ECS for 0.1<AOD<0.3 and over the YRD for AOD>0.3. Statistically significant data points are indicated with * (p value < 0.01).

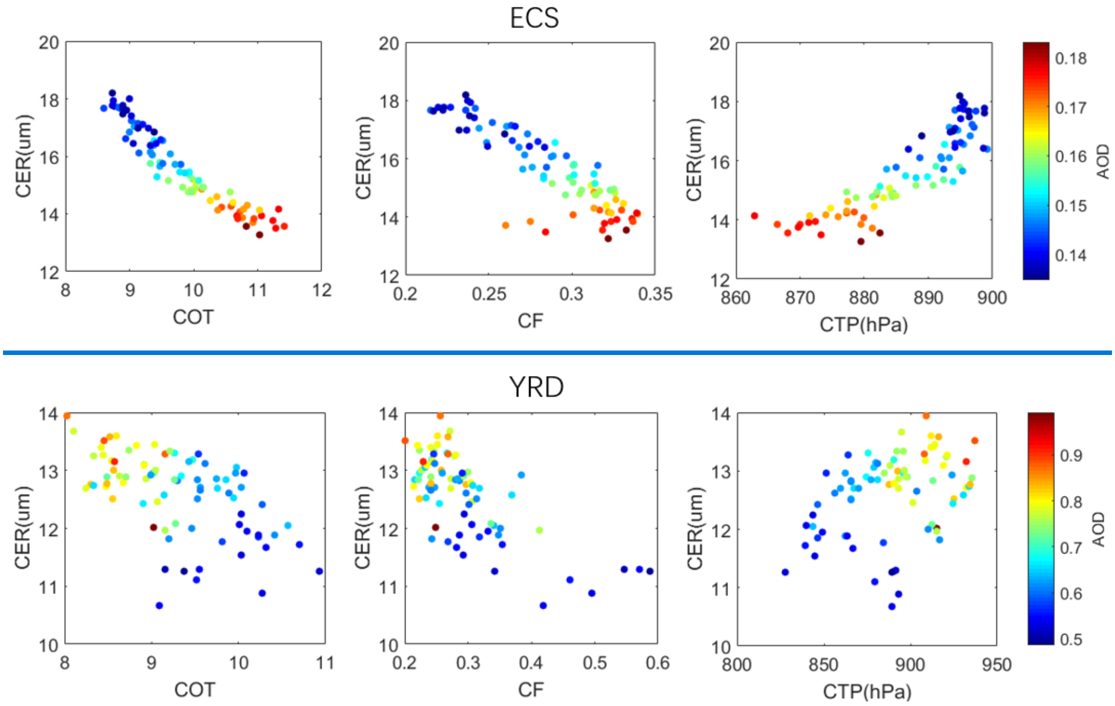
	~~ECS (0.1<AOD<0.3)~~		~~YRD (AOD>0.3)~~	
~~LWP~~	~~ACI~~	~~R~~	~~ACI~~	~~R~~
~~0-40~~	~~-0.13~~	~~0.99*~~	~~-0.12~~	~~0.91*~~
~~40-80~~	~~0.17~~	~~-0.98*~~	~~-0.13~~	~~0.96*~~
~~80-120~~	~~0.35~~	~~-0.99*~~	~~-0.10~~	~~0.76*~~
~~120-160~~	~~0.41~~	~~-0.99*~~	~~0.01~~	~~-0.10~~
~~160-200~~	~~0.42~~	~~-0.99*~~	~~0.08~~	~~-0.37*~~

	ECS (0.1<AOD<0.3)		YRD (AOD>0.3)	
LWP (g m^{-2})	~~ACI~~S	R	~~ACI~~S	R
0-40	-0.10	0.94*	-0.08	0.63*
40-80	-0.19	-0.98*	-0.10	0.81*
80-120	-0.38	-0.99*	-0.06	0.57*
120-160	-0.41	-0.99*	-0.03	-0.11
160-200	-0.46	-0.98*	-0.14	-0.42*

4.4 Behaviour of CER and other cloud properties with the increase of AOD

Scatterplots of ~~the~~ CER versus other cloud properties (COT, CF and CTP), with AOD as third parameter (color-coded)~~, are presented in Figure 8~~, over the ECS and the YRD, are presented in Figure ~~8~~7. Over the ECS, the CER and CTP decrease (the cloud top height increases) with the increase of AOD, and the COT and CF increase. The increase of AOD indicates an increase of the aerosol concentration and thus potentially the number of CCN, which in turn, upon activation, results in the increase of the number of cloud droplets and thus an increase of the COT. The positive correlation between COT and AOD over the ECS suggests that the thicker clouds contain more water droplets and are formed in a more polluted atmosphere, which, as discussed in Section 4.2, results from the influence of long-range transport of aerosol produced over land on the aerosol burden over ocean. But at the same time, as F~~f~~igure~~.~~ ~~8~~7(a) shows, CER decreases with increasing AOD, resulting in the increase in cloud albedo and thus also in the increase of COT. The increase of cloud top height with AOD indicates that both the horizontal and vertical expansion of the clouds are also enhanced. These observations are in agreement with the strong correlation between aerosol loading and cloud vertical development for convective clouds over the North Atlantic reported by Koren et al. (2005).

In contrast to the situation over the ECS, over the YRD the increase of AOD results in an increase of the CER and CTP (the cloud top height decreases), and a decrease of the COT. These observations are consistent with those proposed by Liu et al. (2017) in the same study region. The decrease of the CF with increasing AOD could be explained as follows. Due to the high concentration of smoke particles over the YRD (Shen et al., 2021), aerosol particles absorb solar radiation which results in local heating of the aerosol layer and cooling of the surface (Li et al., 2017). This in turn stabilizes the temperature profile and reduces the relative humidity and surface moisture fluxes (evapotranspiration) (Koren et al., 2008) and thus also cloudiness. Reduced cloud cover exposes greater areas of the aerosol layer to direct irradiation from the Sun and therefore produces more intense heating of the aerosol layer, further reducing cloudiness (Koren et al., 2008). It is noted that this process is different from that proposed by Liu et al. (2017), i.e. that the CF increases with increasing AOD in polluted and heavily polluted conditions (AOD>0.3). In the study of Liu et al. (2017), the LWP range was not constrained, i.e. ~~the~~ aerosol-cloud interaction ~~relationship~~ was studied considering the whole LWP range. The data presented in Table ~~2~~3, shows that ~~the ACI~~S significantly changes between ~~different LWP regimes, i.e. for~~ the three LWP intervals between 0 and 120 g m^{-2}~~,~~ ~~the~~ where ~~ACI~~ S is ~~negative~~ positive (anti-Twomey effect) and for larger LWP it is ~~positive~~ negative but statistically not significant. Figure ~~9~~ 8 shows that CER and CTP substantially increase, whereas COT and CF decrease with increasing AOD in the two LWP intervals between 40-120 g m^{-2}. However, in the other three LWP intervals the relationships between these cloud parameters and AOD are not evident. The different explanations offered here and in Liu et el. (2017) may be related to the different aerosol and cloud ~~properties~~ data sets used by Liu et al.~~,~~ (2017) and in the current study. On the one hand, the data sets have a different spatial resolution and cover a different time period. The dataset used in the study of Liu et al. (2017) are MYD04 Level 2 Collection 5 and MYD06 Level 2 Collection 5 in the period from 2007 to 2010. During that period the AOD over the YRD was at a maximum and decreased substantially in later years (Liu et al, 2021; de Leeuw et al., 2022; 2023). On the other hand, in the study of Liu et al. (2017), the MODIS-retrieved AOD was averaged over an area with a radius of 50 km from the CALIOP target and the MODIS-retrieved cloud data were averaged within a radius of 5 km from the CALIOP target. Hence the AOD and cloud parameters were not representative for the same area, in particular in cases with inhomogeneous spatial distributions.



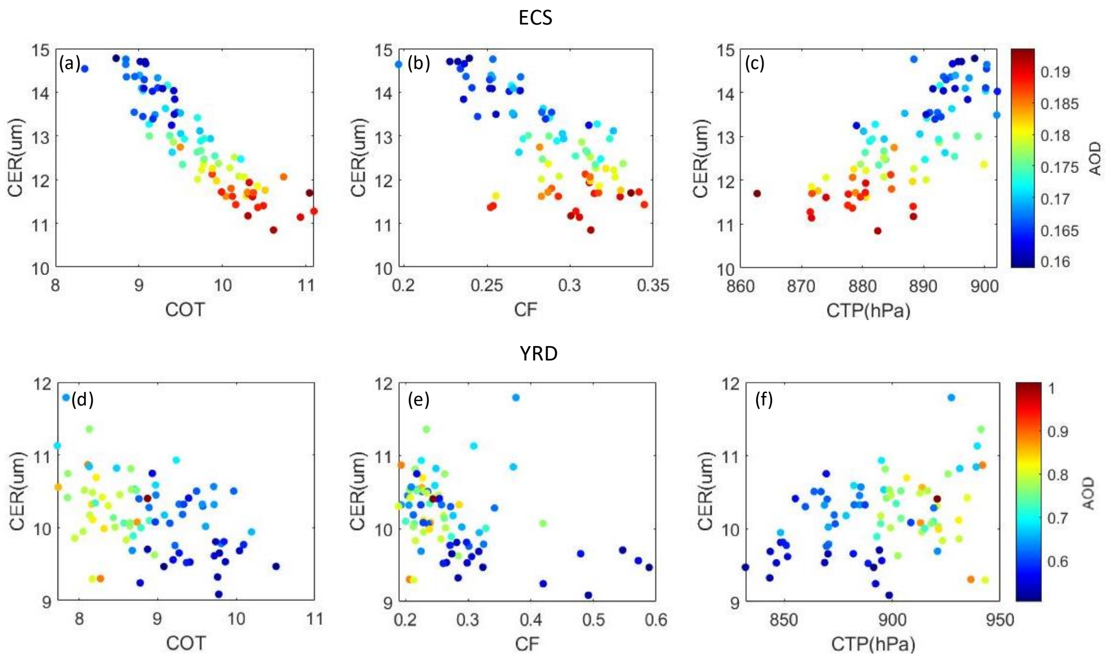
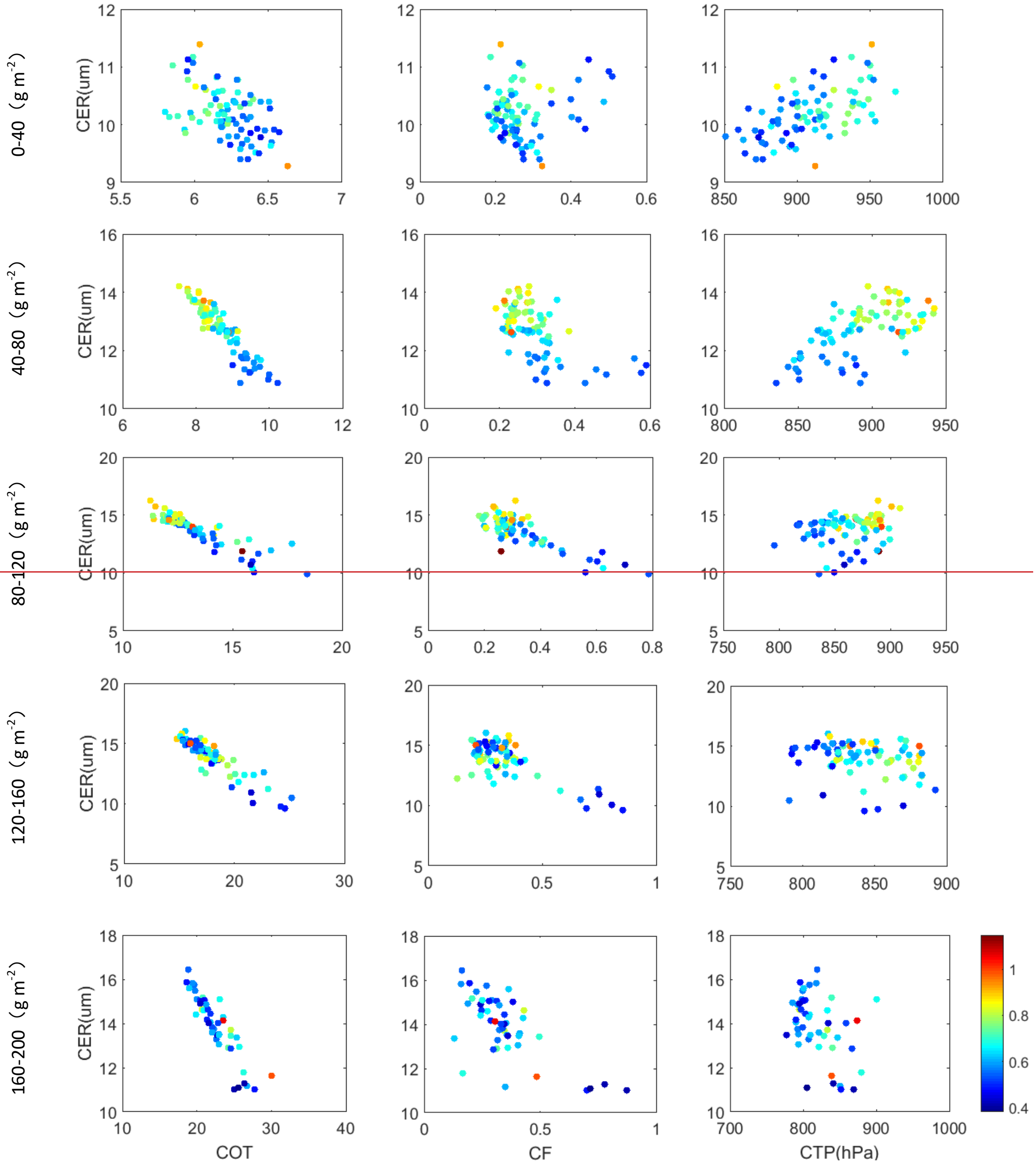


Figure ~~8~~7. Scatterplots of CER versus other cloud parameters (COT, CF and CTP; left to right) over the ECS

(top row) and the YRD (bottom row), with AOD as third parameter, color coded following the scale at the right.



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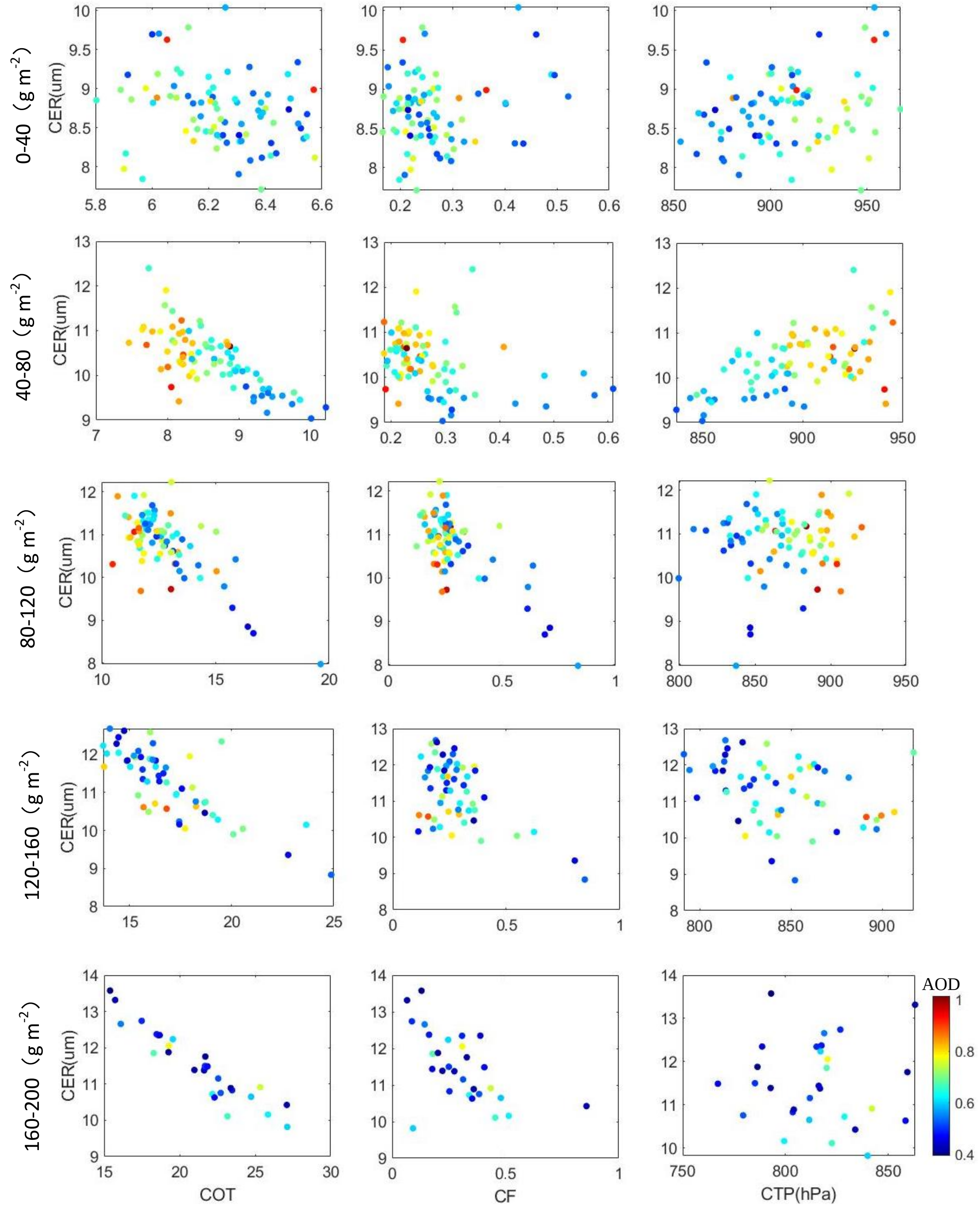


Figure ~~9~~8. Scatterplots of CER versus other cloud parameters (COT, CF and CTP; left to right) over the YRD, for ~~three~~ five different LWP intervals between 0 and 200 gm^{-2}. The AOD for each grid point is color coded following the scale at the right.

4.5 Behaviour of CER and AOD in different meteorological conditions

Scatterplots of the CER versus AOD over the ECS and the YRD, with meteorological factors (LTS, RH, PVV) (color coded) as third parameter, are presented in Figure ~~10~~9. Over the ECS (Figure ~~10~~9(a)), the AOD is inversely related to LTS, whereas the CER increases with increasing LTS. This observation is different from the findings of Saponaro et al. (2017) who reported that there is no significant influence of atmospheric stability (LTS) on the relationship between CER and AOD. Likewise, the AOD is inversely related to RH whereas CER increases with increasing RH. These two observations indicate that RH and LTS have a similar effect on the relationship between AOD and CER. In contrast, with the increase of PVV, the AOD becomes larger but the CER becomes smaller. The CER vs AOD curves show that, overall, the meteorological conditions do not change the functional relationship between AOD and CER, but quantitatively they do have an effect. The change of meteorological conditions plays an important role in the variation of CER. ~~In addition, Figure 10b shows that, over the ECS, with the increase of the aerosol concentrations, the number of cloud condensation nuclei also increases, so the same amount of water vapor is distributed over a larger number of cloud droplets resulting in smaller cloud droplets. Hence, the interaction between AOD and CER over the ECS is in agreement with the Twomey cloud albedo effect.~~

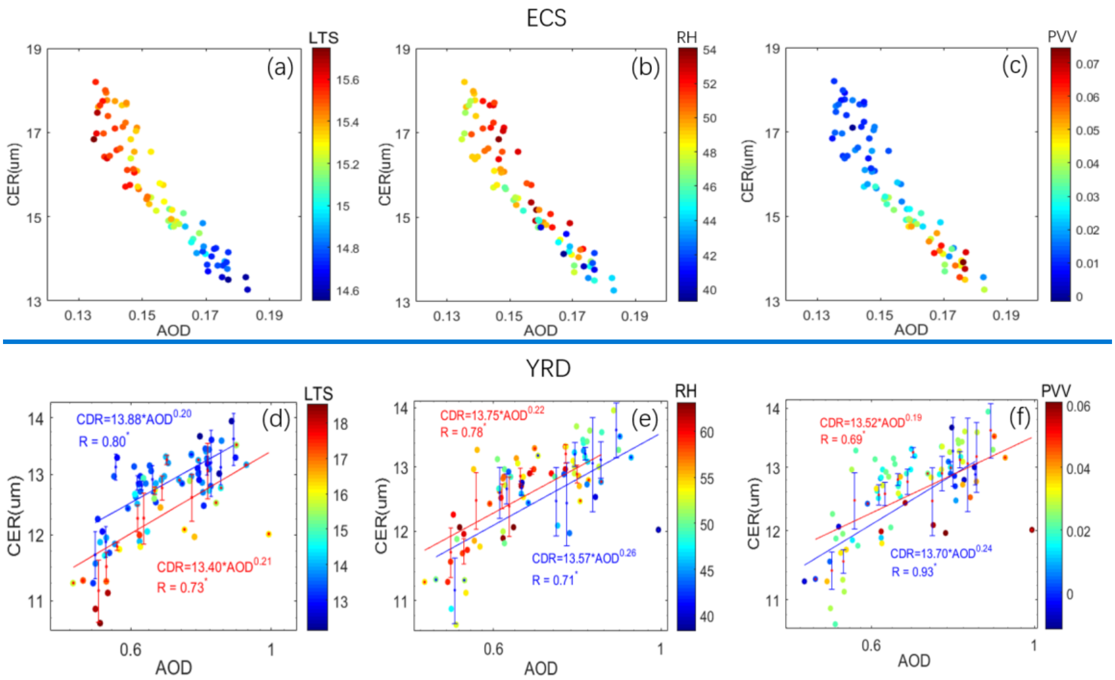
~~Cloud properties as a function of AOD for two different LTS conditions over the YRD are presented in figure 10d, i.e. for low LTS, with a mean value of 13.27 representing an unstable atmosphere; and for high LTS, with a mean value of 15.23 representing a stable atmosphere. In unstable atmospheric conditions the CER is larger than in stable conditions, and in both unstable and stable conditions CER increases with AOD. The larger CER in unstable conditions may be due to better vertical mixing of both aerosol particles and water vapor (Liu et al., 2017).~~

~~The effect of relative humidity (RH, at 750 hPa) on the relationship between the cloud properties and AOD is evaluated by dividing the data into two equally sized subsets for high and low RH, and the mean relative humidity values for each subset are calculated. In high relative humidity conditions (57%) the CER is much larger than in low relative humidity conditions (48%), as shown in Fig. 10e.~~

Different from the situation over the ECS, over the YRD the effect of meteorological conditions on the CER is weak as shown in Figures 9(d)-(f). RH and PVV ~~shows~~ have an inverse ~~role in~~effect on the relationship between AOD and CER. There is no significant influence of atmospheric stability (LTS) on the relationship between CER and AOD as suggested by Saponaro et al. (2017). Overall~~Therefore,~~ aerosol concentration plays a ~~dominant~~more important role in the effects of different factors on CER over the YRD.

~~The effect of vertical velocity (PVV) on the CER in polluted and heavily polluted conditions is weak. In~~

~~general, the influence of aerosol concentration (AOD) on the CER is larger than that of meteorological conditions, although the combined effect of AOD and meteorological conditions is larger than that of AOD alone (Fig. 7).~~ ~~Therefore, aerosol concentration plays a dominant role in the effects of different factors on CER.~~



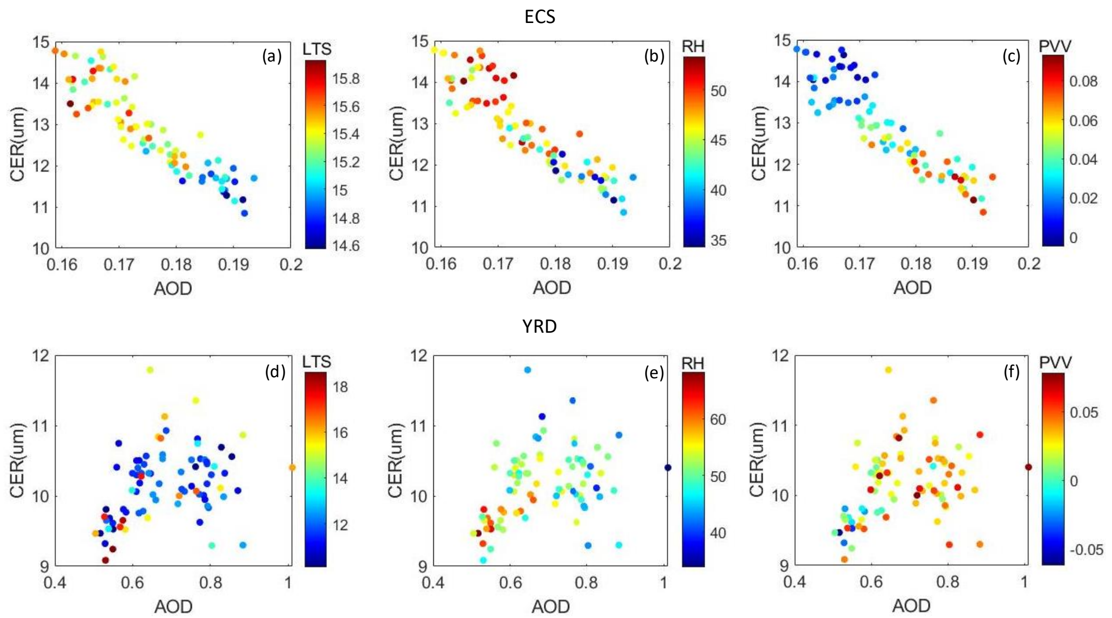


Figure ~~10~~9. Scatterplots of CER versus meteorological parameters (LTS, RH and PVV; left to right) over the ~~E~~CS (top row) and the YRD (bottom row). The AOD for each grid point is color coded following the scale at the right. ~~The lines in the bottom row (YRD) present the least-square fits for low and high LTS, RH and PVV, respectively, as described in the text, and the resulting relations are presented in each figure. The marker *~~

~~**at the top right corner of R value denotes statistically significant if p < 0.05.**~~

4.6 Application of the ~~Geographical~~ geographical detector ~~model~~ method ~~analysis~~

4.6.1 Factor detector analysis

The GDM factor detector module was used to analyze the influence of 9 factors (~~aerosol~~AOD, cloud and meteorological ~~conditions~~parameters) on ~~ACI~~ S over the YRD and the ECS, for the conditions summarized at the end of Section 4.3. These factors are summarized in Table ~~3~~4, together with q, i.e. the explanatory ~~ir~~ power of that factor to S ~~q-values~~ (~~Equation~~ Eq. 2), over the ECS and the YRD. The data in Table ~~3~~ 4 show that the influences of the 9 proxy variables on S are rather wea~~e~~k and not statistically significant. ~~and~~They can explain only 1%-15% of the variation of S in both target regions.

~~over the ECS, only the influences of the proxy variables CER and CF have some are statistically significancet with p < 0.1. Over the YRD, the variables CER, LWP, CF, RH and LTS all exertall have some significant impacts on ACI CER but with small statistical significance (at the p < 0.1) level. In addition to the weak statistical significances, aHowever,lso the q-values show that the influences of the 9 se variables on the ACI valuesS is are rather weak and can explain only 10%-22% of the variation of the ACIS. Furthermore, in neither of the two regions the influence of the proxy variables AOD, COT, CTP and PVV is statistically significant at the 0.1 level.~~

Table ~~3~~4. q values for factors which may influence ~~ACI~~ S over the ECS and the YRD, evaluated for data collected in the period from 2008-2022.

	~~Aerosol parameter~~	~~Cloud parameters~~					~~Meteorology parameters~~		
	~~AOD~~	~~CER~~	~~COT~~	~~LWP~~	~~CF~~	~~CTP~~	~~RH~~	~~LTS~~	~~PVV~~
~~ECS~~	~~0.06~~	~~0.14*~~	~~0.16~~	~~0.10~~	~~0.16*~~	~~0.10~~	~~0.04~~	~~0.04~~	~~0.09~~
~~YRD~~	~~0.06~~	~~0.10*~~	~~0.05~~	~~0.22*~~	~~0.21*~~	~~0.05~~	~~0.13*~~	~~0.16*~~	~~0.04~~

Study Area	Aerosol parameter	Cloud parameters					Meteorologica~~l~~y parameters		
	AOD	CER	COT	LWP	CF	CTP	RH	LTS	PVV
ECS	0.07	0.06	0.06	0.10	0.01	0.13	0.10	0.11	0.09
YRD	0.05	0.09	0.06	0.05	0.04	0.06	0.15	0.09	0.09

Note: *** indicates that the q value is significant at the 0.01 level (p < 0.01).

The GDM factor detector module was also used to analyze the influence of the AOD and meteorological parameters ~~factors~~ (RH, LTS and PVV) on adjustments of cloud properties. The results in Table ~~4~~ 5 show that ~~over the ECS,~~ AOD~~, LTS~~ and PVV influence all cloud parameters over the ECS except CTP, with q-values which are statistically significant at the 1% level. The ~~high~~ q-values for AOD show that this factor ~~e AOD~~ can explain 46~~71~~% (for CF) to 81~~7~~% (for CER) of the variation in the cloud parameters considered in this study, and PVV can explain 47% (for CF)~~and 47% (for COT), which both are similar to the explanatory power of AOD,~~ to 70% (for CER) of the variation in the cloud parameters. For LTS and RH, the q-values for CER are statistically significant but with smaller explanatory power than for AOD and PVV. In contrast, the q-value of LTS for LWP is statistically significant and not much smaller than for PVV. ~~which is substantially more than the explanatory power of the meteorological parameters. Among the meteorological parameters, LTS has the largest influence on cloud parameters and the effect of RH is the smallest.~~

Table ~~4~~5. q values for factors which may influence cloud parameters over the ECS, evaluated for data collected in the period from 2008-2022.

~~—~~	~~AOD~~	~~RH~~	~~LTS~~	~~PVV~~
~~CER~~	~~0.87***~~	~~0.36***~~	~~0.75***~~	~~0.69***~~
~~COT~~	~~0.83***~~	~~0.48*~~	~~0.79***~~	~~0.69***~~
~~LWP~~	~~0.74***~~	~~0.23~~	~~0.58***~~	~~0.56***~~
~~CF~~	~~0.71***~~	~~0.24~~	~~0.56***~~	~~0.54***~~
~~CTP~~	~~0.78~~	~~0.54~~	~~0.70~~	~~0.74~~

Cloud parameters	**AOD**	**RH**	**LTS**	**PVV**
CER	0.81***	0.33***	0.44***	0.70***
COT	0.69***	0.40	0.38	0.67***
LWP	0.68***	0.23	0.43***	0.49***
CF	0.46***	0.20	0.09	0.47***
CTP	0.47	0.53	0.18	0.58

Note: ***indicates that the q value is significant at the 0.01 level ($p < 0.01$). ~~** indicates that the q value is significant at the 0.05 level (p < 0.05). * indicates that the q value is significant at the 0.1 level (p < 0.1).~~

The results from a similar analysis of the data over the YRD (Table ~~5~~6) show that AOD has a statistically significant influence at the 1% level on COT and CF, but with much smaller explanatory power than over the ECS. AOD can explain 31~~54~~% of the variation of CER but the statistical significance is small ($p<0.1$).

Among the meteorological parameters, RH has a statistically significant influence on CTP and can explain 74~~65~~% of the variation of the CTP and LTS can explain ~~48~~55% of the variation of the LWP and 50% of the variation of the CF with p<0.01. The explanatory power for the effects of RH (~~37~~32%)~~, LTS (46%)~~ and PVV (18~~46~~%) on LWP ~~are substantial but with~~have low statistical significance (p<0.1).

Table ~~5~~6. q values for factors which may influence cloud parameters over the YRD, evaluated for data collected in the period from 2008-2022.

~~—~~	~~AOD~~	~~RH~~	~~LTS~~	~~PVV~~
~~CER~~	~~0.54*~~	~~0.05~~	~~0.41~~	~~0.14~~
~~COT~~	~~0.61***~~	~~0.27~~	~~0.04~~	~~0.19~~
~~LWP~~	~~0.18~~	~~0.37*~~	~~0.46*~~	~~0.46*~~
~~CF~~	~~0.33***~~	~~0.04~~	~~0.48***~~	~~0.12~~
~~CTP~~	~~0.48~~	~~0.65***~~	~~0.22~~	~~0.38~~

Cloud parameters	**AOD**	**RH**	**LTS**	**PVV**
CER	0.31	0.25	0.13	0.18
COT	0.61***	0.45	0.12	0.29
LWP	0.16	0.32	0.55***	0.18
CF	0.30***	0.02	0.50***	0.07
CTP	0.50	0.74***	0.32	0.56

Note: ***indicates that the q value is significant at the 0.01 level (p < 0.01).

~~** indicates that the q value is significant at the 0.05 level (p < 0.05). * indicates that the q value is significant at the 0.1 level (p < 0.1).~~

~~In t~~Tables 5 and 6 list q values ~~were given~~ for individual factors, together with p showing the absence of statistical significance in many cases, especially over the YRD, and often the explanatory power is not ~~so~~ high when ~~low~~the significance is low. These data show ~~We find~~ that cloud parameters ~~is~~are ~~seem to be~~ dominated by aerosol effects over the ECS but meteorological influences on cloud parameters predominate over the YRD, as was also concluded from the analysis from “traditional” statistical methods presented in Section 4.5 and these conclusions are~~which is~~ consistent with the results published by Andersen and Cermak (2015). Among the meteorological parameters, we also find that PVV (with highest q in the three meteorological parameters) predominantely influences~~ on~~ cloud parameters over the ECS. Jones et al. (2009) and Jia et al. (2022) reported that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. In addition, we find that CTP is mainly affected by RH ($q = 0.74^{***}$) and PVV ($q = 0.56$) over the YRD, as suggested by Koren et al. (2010). Koren et al.~~The author~~ reported that observed cloud top height correlates best with model pressure updraft velocity and relative humidity. To some extent, LTS influences ~~on~~ CER (q = 0.44***) and LWP (q = 0.43***) over the ECS, while, in contrast, over the YRD LTS predominately influences ~~on~~ CF (q = 0.50***) and LWP (q = 0.55***)~~ over the YRD~~. Matsui et al. (2004) and Tan et al. (2017) reported that aerosol impact on CER is stronger in more dynamic environments that feature a lower LTS and argue that very high LTS environments dynamically suppress cloud droplet growth and reduce aci intensity. While strong correlations between AOD and cloud parameters have been previously observed, they are likely due to the swelling of aerosol particles in humid airmasses (Quaas et al, 2010), rather than an aerosol influence, which is in agreement with findings by, e.g., Myhre et al. (2007), Twohy et al. (2009) and Quaas et al. (2010).

4.6.2 Interaction detector analysis

The q values of the combined effect of two parameters (AOD, RH, LTS, PVV) ~~interactive q-statistic values for the~~ influenc~~e~~ing~~ of AOD and meteorological parameters on~~ the cloud parameters over the YRD and the ECS, derived using the GDM as described in Section 3.2, are ~~graphically~~ presented in the matrix ~~shown~~ in Figure ~~7~~10~~, with the q-values color coded~~. The data in Figure ~~7~~ 10 show that the ~~interactive~~ q-values for the interaction of a pair of factors are larger than the q-values for any of the individual parameters (Table ~~4~~5). Over the ECS, the combined ~~influences~~ effects all exhibit a ~~binary nonlinear~~bilinear ~~–~~enhancement over the time period of this analysis. ~~Calculations show that t~~The q values for the combined effects on CER over the ECS show that ~~of AOD and RH on the CER results in~~ the ~~highest~~ explanatory power of AOD together with each of the three meteorological parameters, RH, LTS and PVV is high with~~92%,~~ 86%, 84% and 9~~1~~2%, respectively~~as indicated by the q value (color bar at the bottom)~~. Also for the combination of LTS and PVV the explanatory power is high (90%). Further inspection of the data in Figure ~~7~~ 10 shows that the explanatory powers of the combined effects are high for several combinations of parameters, such as ~~for CER combining AOD with LTS or PVV, and LTS with PVV,~~ for COT the combination of AOD with RH, LTS or PVV~~ or LTS with PVV, etc~~. The data in Figure ~~7~~ 10 show that the combination of AOD and ~~RH~~ PVV results in high explanatory power for their influence on ~~all 5~~4 cloud parameters (CER, COT, LWP and CF) and the combination of LTS with ~~PVV~~ RH has high explanatory power for their effects on ~~CER, COT, LWP and~~ CTP. Among the meteorological parameters, we find that the combined effect of AOD and PVV predominantely influences ~~on~~ cloud parameters over the ECS. The result is in accord with the findings of Jones et al. (2009) and Jia et al. (2022) that stronger aerosol cloud interactions typically occur under higher updraft velocity conditions. Over the YRD, half of the ~~interactions~~ q values for the combined effects on cloud properties exhibit nonlinear enhancement ~~of the influence of the independent parameters on the cloud properties~~ over the time period of this analysis, in~~ferring~~dicating that the combined effects on cloud properties are much larger than that over the ECS. The data in Figure ~~7~~10 show that the combination of AOD and RH results in high explanatory power for their influence on CER and COT, and the combination of AOD with LTS has high explanatory power for their effects on LWP and CTP. ~~The results in Figure 7 10 show that t~~The combined effects of ~~AOD~~ PVV and LTS on the CF result in the highest explanatory power of ~~0.70~~0.84. The data in Fig. ~~7~~10 also show that the explanatory power is largest for the combined influence of AOD together with other factors, and is somewhat larger than the influence of AOD alone (Table ~~5~~6) for all 5 cloud parameters. ~~To some extent, this also applies to RH.~~ What's more, the data do show that meteorological factors enhance the explanatory power of the ~~cloud factors~~AOD on cloud parameters over both ~~two~~ regions.~~;~~ ~~for~~ For example, the individual q values for the influence of AOD and PVV over the ECS were 0.~~83~~81 and ~~0.69~~0.70 but for the combined influence the q-statistic is as high as 0.~~90~~92. ~~May be we need to write more here~~The results from the GDM interaction detector analysis clearly show the enhancement of the interaction q-values over the q-values for the individual factors. In other words, the explanatory power of the combined effects of aerosol and a meteorological parameter ~~and aerosol~~ is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters. We can conclude that aerosol and meteorological conditions do make a significant contribution to cloud parameters and that confounding effects of different factors are often more important than each parameter alone and that the relative importance of each parameter differs significantly over the ECS and YRD.

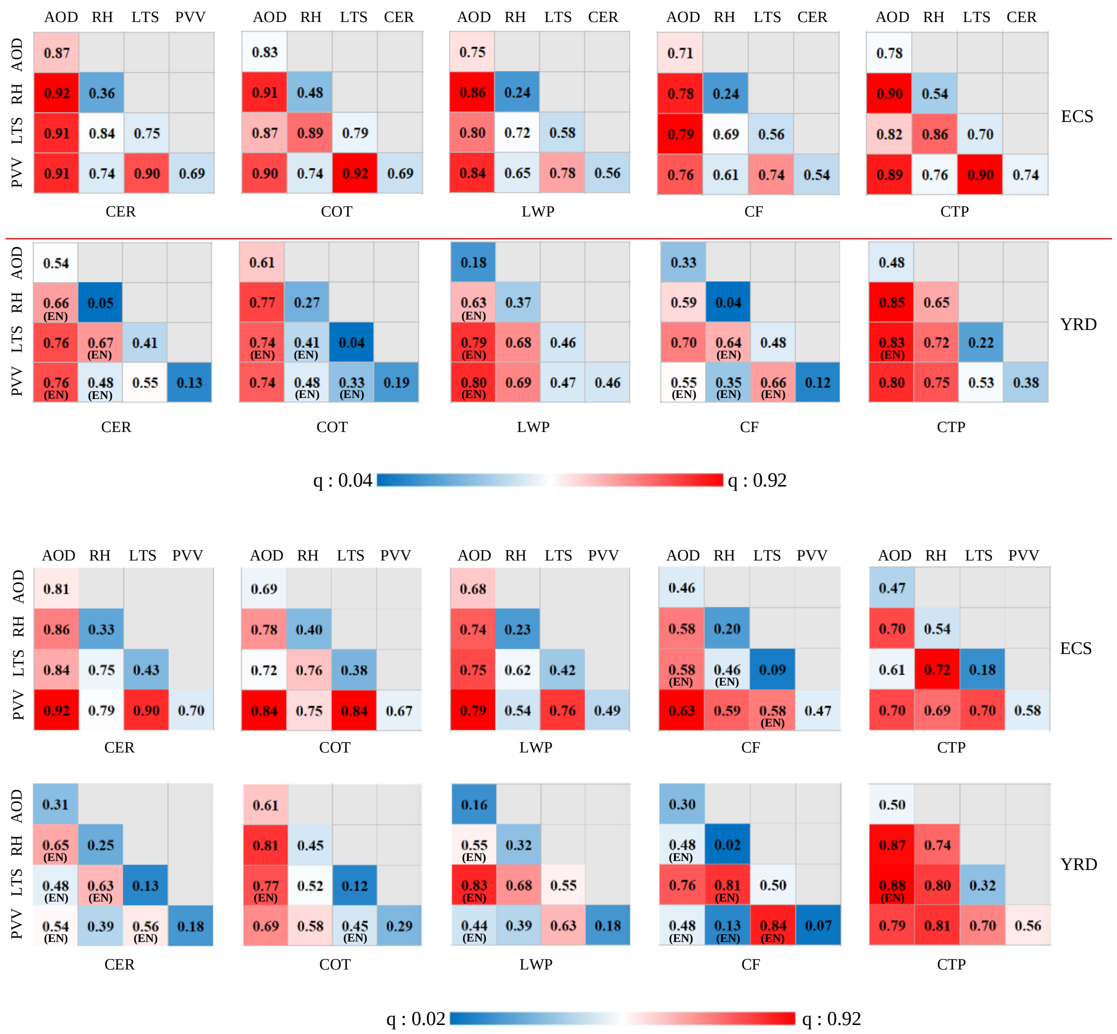


Figure ~~7~~10. q values derived using the GDM for the combined effects of AOD, RH, LTS and PVV on cloud parameters ~~Interactive proxy variable q statistics~~ over the ECS (top) and the YRD (bottom). In addition to the numbers, the q values are colour coded according to the colour scale (linear from 0.04 to 0.92) at the bottom, for easy identification. ~~Note:~~ (EN) below a q value indicates the nonlinear enhancement of two variables (if $q(x1 \cap x2) > q(x1) + q(x2)$), the absence of a ~~and no~~ label below a q value ~~denotes the~~indicates a ~~binary~~ bilinear enhancing of two variables (if $q(x1 \cap x2) > Max[q(x1), q(x2)]$) ~~(Wang and Hu, 2012)~~.

5 ~~Conclusions~~Discussion

Warm cloud properties over eastern China have been investigated in relation to aerosol and meteorological conditions using 15 years (2008-2022) of data from passive (MODIS/Aqua) satellite measurements, together with the ECMWF ERA-5 reanalysis ~~data~~~~ERA Interim Reanalysis~~ meteorological data. The Yangtze River Delta, a heavily polluted region in eastern China, and the East China Sea with a relatively clean atmosphere, were selected as study areas. Relationships between cloud droplet effective radius and AOD (used as a proxy for ~~aerosol concentration~~CCN), i.e. ~~characterized by~~ the sensitivity S of CER to changes in AOD~~aerosol cloud interaction (ACI) index~~, were constructed for different constraints of AOD and LWP. The effects of AOD on CER were investigated for three AOD regimes. In view of the uncertainty of MODIS-retrieved AOD and the scatter in the CER-~~/~~AOD relations, data for AOD<0.1 were not considered~~ in this study~~. In the moderately polluted AOD regime (0.1<AOD<0.3)~~AOD regime, with 0.1 to 0.3~~, the CER over the YRD did not change significantly with AOD, whereas over the ECS the CER strongly decreased with AOD and the derived relationship between CER and AOD is statistically significant. In the third AOD regime, with AOD > 0.3, ~~over the YRD~~ the CER increased with increasing AOD over the YRD. In contrast, over the ECS there was no clear relation between CER and AOD, although CER variability increased with AOD>0.3, especially for higher AOD (> ~0.8)~~, whereas over the ECS the CER did not significantly change as function of AOD, although the variations in the CER increased with AOD, especially for higher AOD (> ~0.8)~~. Based on these results, two different AOD regimes were selected for further investigation of ~~the ACI~~aci: 0.1<AOD<0.3 over the ECS and AOD > 0.3 over the YRD. The spatial distribution of S~~ACI~~, here defined as the relative change in CER as a function of the relative change in AOD (~~eq~~Eq. 1), averaged over the 15-years study period, shows that it was negative and statistically significant over the ECS and positive over the YRD. These results were obtained using data with no restriction on LWP. Stratification by ~~Further selection of the data in different~~ LWP ~~intervals~~ shows that over the YRD, for AOD >~~greater than~~ 0.3, ~~ACI~~ S is ~~negative~~ positive for LWP in the interval [0-120 g m^{-2}] with very small differences between three LWP intervals (0-40, 40-80 and 80-120 g m^{-2}). In contrast, over the ECS, for AOD in the range from 0.1 to 0.3, ~~ACI~~S is ~~positive~~ negative in the LWP interval [40-200 g m^{-2}] and the value of S is ~~re are~~ substantially ~~differences~~ different between the 4 LWP intervals, with ~~ACI~~ S increasing with LWP, as shown in Table ~~2~~3.

These results were obtained using data from a period of 15 years. During this period, the aerosol properties changed in response to expanding economy, resulting in the increase of the AOD until 2007, and the implementation of emission reduction policy resulting in the decrease of the AOD from 2014 which flattened from about 2018 (de Leeuw et al., 2021; 2022; 2023). To account for these changes, the sensitivity S was determined for the periods 2008-2014 and 2014-2022, without stratification for LWP (see Figures xx ~~FF: the 4 figures in the response~~in the S~~s~~upplementary). The results for the ECS show no significant difference between the CER-AOD relations during these two periods. Over the YRD, however, the data for 2008-2014 show a clear decrease of CER with increasing AOD for 0.1<AOD<0.3

and for larger AOD the CER increased, with a statistical significant correlation (R=0.87) and S=0.10 as compared to S=0.08 for the whole period. In contrast, the data for 2014-2022 show no clear correlation between CER and AOD for both AOD intervals over the YRD. A similar exercise for shorter periods, i.e. for each year between 2008 and 2022, show similar behavior as for the whole period 2008-2022, over both study areas, with interannual variations of the value of S. However, the statistical significance is low (large p) due to the small number of data samples in each year.

It is noticed that in recent papers (e.g., Gryspeerdt et al., 2023; Arola et al., 2022) the usefulness of correlating aerosol and cloud parameters has been seriously challenged because cloud variability and retrieval errors are such that correlations between ~~aerosol optical depth~~AOD and cloud properties (N_d, CER, LWP) can be spurious. Gryspeerdt et al. (2023) discussed ~~ACI~~aci in terms of the susceptibility β of N_d to aerosol rather than the sensitivity S of CER to aerosol (see the discussion in the Introduction on the use of N_d vs CER), and the problem arises with low aerosol conditions due to larger aerosol retrieval uncertainty due to surface correction (larger surface effect on the radiance at the top of the atmosphere), which applies equally to β and S. In the current study we did not consider the lowest aerosol conditions by limiting the data to situations with AOD ≥ 0.1, as discussed in Section 4.2. Furthermore, we stratified the analysis for moderate (0.1 ≤ AOD < 0.3) and high (0.3 ≥~~≤~~ AOD) aerosol regimes, based on the data.

Arola et al. (2022) addressed the susceptibility of N_d to changes in aerosol and the adjustment of LWP (using satellite observations), and confounding factors, in particular co-variability of N_d and LWP induced by meteorological effects. They show how errors in the retrieved CER and COT or spatial heterogeneity in cloud fields influence the N_d - LWP relation. However, both N_d and LWP are not retrieved but derived from CER and COT. Using Eq. 1 and Eq. 2~~eqs. 1 and 2~~ in Arola et al. (2022), the N_d-~~/~~LWP relationship can be shown to have a highly non-linear dependence on CER and thus it is no surprise that any error in CER strongly affects the relation between N_d and LWP. Their experiments, i.e. using smaller scales (5° x 5°) to reduce spatial meteorological variability, or using snapshots to remove meteorological variability in time, did not lead to a conclusion whether the N_d - LWP variability is due to spatial heterogeneity in the cloud fields or due to retrieval errors. The main message from this part of the study (using satellite data) by Arola et al. (2022) is "the spatial variability of CER introduces a bias which moreover becomes stronger in conditions where the CER values are lower on average".

Experiments with simulated measurements show that "the main cause of the negative LWP vs N_d slopes is the error in CER". Arola et al. emphasize that the spatial cloud variability and retrieval errors in CER and COT are similar sources for negative bias in LWP adjustment and that these sources could not be separately assessed in their simulations. The implication of the findings of Arola et al. (2022) on the adjustment of LWP for the results of the current study on the sensitivity of CER to aerosol (or CCN, using AOD as proxy) is that the assumption of constant LWP may be violated. This would affect the results presented in Section 4.3 where LWP was stratified and S was found to vary with LWP. In view of the LWP adjustment to changes in aerosol, the variation of CER sensitivity with LWP may be somewhat different from that reported in section 4.3.

The above results were obtained by using traditional statistical methods where relationships were derived from scatterplots of CER versus AOD, stratified in two different AOD regimes and five different LWP regimes, as discussed above. The data were also analyzed by ~~By~~ using the ~~geographical detector method~~GDM to determine which factors influence ~~ACI~~aci and identify how interactions between different parameters influence the results of the aci analysis, i.e. the sensitivity and resulting adjustments. In particular, the GDM provides information on the extent to which the effect of individual factors is influenced by other factors. As shown in Section 4.6.1, the effect of individual factors may be overestimated when confounding effects of other factors are not accounted for. The interaction detector analysis (Section 4.~~2~~6.2) shows a more realistic estimate of the effects on aci when different factors are analyzed together. The factor detector analysis (Section 4.6.1) shows that ~~, the influence of different parameters (AOD and meteorological parameters) on the cloud properties in eastern China has been investigated. O~~over the ECS, AOD has the largest influence on cloud parameters, as indicated by the large and statistically significant q values. Among the meteorological factors, ~~LTS~~ PVV has more influence on the variations of the cloud parameters than RH and ~~PVV~~LTS. Over the YRD, AOD has the largest influence on ~~CER and~~ COT, with large and significant q values. Among the meteorological factors, the effect of LTS on CF is greater than that of RH and PVV. However, the ~~the~~ q-values may sum up to over 100% when the variables are not independent, i.e. the explanatory power of such variables is too high. The evaluation of the effects of interaction between different factors on aci corrects these clearly unrealistic situations. The analysis in section 4.~~2~~6.2 shows that the interactive q-statistic values derived in this study ~~were~~ are larger than any of the values for single variables, i.e. the explanatory power of a combination of factors is higher than that of individual factors, but less than 100%. ~~The combined influences of AOD and meteorological parameters exhibit binary nonlinear enhancement of the explanatory power of the variation of the cloud parameters.~~ However, although the GDM~~this work can therefore only~~ provides ~~further~~ evidence of the effects of aerosol and meteorological factors and their interactions ~~effect~~ on cloud propertiess and quantify the relative contributions~~and combined effects on clouds~~ to aci, it ~~but~~ cannot quantify the absolute contributions with confidence.

6 Conclusions

The response of different cloud parameters to variations in AOD and in meteorological conditions has been analyzed using traditional statistical methods to determine the sensitivity S of CER to aerosol for different aerosol regimes and stratified according to LWP. The results show the contrasting behavior over a polluted region over land (YRD) and a relatively clean region over ocean (ECS). In the intermediate aerosol regime ($0.1<AOD<0.3$), CER does not significantly change with AOD over the YRD ($S\approx0$), but over the ECS S is negative and increases with increasing LWP. In the high aerosol regime ($AOD>0.3$), S is positive over the YRD but varies little with LWP, whereas over the ECS the CER does not change with AOD. These results may be influenced by confounding effects of meteorological parameters. The study further shows that over the ECS the CER is larger for higher LTS and RH but lower for higher PVV. Over the YRD, there is no significant influence of LTS on the relationship between CER and AOD.

The GDM has been applied to determine which factors influence S and cloud parameters and the interaction detector analysis has been used to determine the combined effect of different parameters on cloud parameters. ~~The results show that o~~~~Over the ECS, with the increase of the AOD, the CER and CTP decrease, and the COT and CF increase. Over the YRD, with the increase of the AOD, the CER and CTP increase, and the COT and cloud cover decrease. The CER is larger in unstable atmospheric conditions than in stable conditions, irrespective of the AOD. The cloud fraction is much larger in high relative humidity~~RH ~~conditions than in low relative humidity~~RH ~~conditions. However, the impact of vertical velocity~~PVV ~~on the CER is weak in polluted and heavily polluted conditions. In general, the influence of the AOD on the CER is greater than that of meteorological conditions. Therefore, AOD plays a dominant role in the effects of different factors on CER.~~

The results from the GDM interaction detector analysis clearly show the enhancement of the interaction q-values over the q-values for the individual factors. In other words, the explanatory power of the combined effects of aerosol and a meteorological parameter is larger than that of each parameter alone. Thus, the GDM provides an alternative way to obtain information on confounding effects of different parameters. We conclude that aerosol and meteorological conditions significantly influence cloud parameters and that combined effects of different factors are often more important than the effect of each individual factor. The relative importance of each factor differs significantly over the ECS and YRD.~~Different from previous studies, here the interaction between aerosol and CER has been investigated by considering different AOD and LWP regimes on ACI over land and ocean. The relative importance of AOD and meteorological parameters on cloud properties were examined by using the geographical detector method.~~

The results of this study contribute to improve the understanding of the indirect effects of aerosols and the role of various driving factors on the cloud microphysical properties. By compari~~ng~~son with aerosol and cloud observation~~al~~s~~data of aerosols and clouds~~, the regional climate model's ability to simulate changes in cloud parameters can be evaluated. A more accurate description of the relative contribution of meteorological factors can improve the parameterization scheme of the model over eastern China.~~They provide a reference for improving the parameterization scheme of regional climate models in eastern China.~~

Data availability

All data used in this study are publicly available. The satellite data from the MODIS instrument used in this study were obtained from https://ladsweb.nascom.nasa.gov/search/ (last access: 12 July 2022, Liu, 2022a). The the ECMWF ERA-5 reanalysis data were collected from the ECMWF ERA-5 reanalysis data server https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels-monthly-means?tab=form (last access: 12 July 2022, Liu, 2022b).

Author contributions

YL and GL designed the research. YL led the analyses. YL and LT wrote the manuscript with major input from JH, GL and further input from all other authors. All authors contributed to interpreting the results and to the finalization and revision of the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant No. 42001290), and the Natural Science Foundation of China (Grant No. 41871253). We are grateful for the easy access to MODIS data products provided by NASA. We also thank ECMWF for providing daily ERA-5 ~~Interim~~ reanalysis data. The study contributes to the ESA / MOST cooperation project DRAGON5, Topic 3 Atmosphere, sub-topic 3.2 Air-Quality.

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