Reviewer 1

1. After reviewing the revised manuscript by Che et al., I regrettably have to recommend not accepting it for publication at ACP. Many of the issues found during the initial review have not been properly addressed.

Response: We thank the reviewer of his or her constructive comments and suggestions. A major overhaul of the paper has been done to address the review comments—all concerns raised in the initial reviews have been considered this time. To this end, the abstract, results analysis, discussion, and conclusion sections all have been rewritten. In the Introduction, the importance of the study has been explained, and the objectives clearly stated. In the section 2, as the reviewer suggested, additional information has been added about the uncertainties and methods of the aerosol retrieval and ARF calculations, and links are now provided to previous studies and literature. Section 3 now presents a more detailed and coherent analyses of the results, and the discussion has been revised and expanded to make the paper more compelling. In the section 4, the conclusions have been rewritten, and they now concisely summarize the most important findings of our study. The detailed issues have been highlighted (in RED) in the revised manuscript.

2. The revised manuscript has plenty of redundancy and poor logic, and fails to present a solid and interesting analysis.

Response: The authors have thoroughly re-organized this manuscript. The sections concerning diurnal AOD cycles, aerosol complex refractive index, comparisons to MODIS results, type classification by EAE and the spectral difference have been deleted.

The flow of the revised manuscript is more straightforward than in previous versions. In brief, we first use data from a dense network composed of seven urban and rural sites to characterize the climatology of aerosol properties (including microphysical and optical characteristics) over the Yangtze Rive Delta (YRD) region. The aerosol properties were then used to calculate direct aerosol radiative forcing (DARF) under clear conditions over the region. As the DARF is affected by aerosol absorptivity as well as the underlying surface conditions, we classified the aerosols by type (strength of absorption for particles in both fine and coarse modes) based on the optical properties. In summary, the results of this study will be of interest to those who study aerosol-climate interactions or air-pollution. The above description summarizes what we have done to improve the flow of the paper.

A more in-depth analysis has been done in the revised manuscript to make the study substantial and interesting. Previous ground-based studies in the YRD have usually been conducted over short or discontinuous periods of time (information on seasonal but not monthly averages has been provided), and therefore information on the temporal and spatial variations in aerosol characteristics over YRD is limited. In this study, we uncovered some important information on aerosol-climate connections through careful analysis of the data.

In brief, (1) we first considered aerosol microphysical properties (particle radius and volume size distributions) and found that the aerosol populations over the YRD region differ from those in northern and northeastern China. High volumes and effective radii of fine-mode aerosols occurred in June and September, and they were consistently lower in July and August. In contrast, high volumes and effective radii of fine-mode aerosol typically occur in July over northern China and northeastern China. This difference can be attributed to the unique regional climatology of aerosols over YRD as we discuss in the revised paper.

(2) Next, the aerosol optical properties over the YRD were analyzed in detail. Aerosol extinction (AOD) also showed two peaks, one in June and the other in September, and a minimum occurred in July because of the fine-mode particle effects. This is unlike northern and northeastern China where only one peak in AOD typically occurs from June to August. Also, the AOD seasonal pattern over YRD in this study is different from the “high in summer but low in winter” pattern in urban areas of north China found in previous studies. The AOD in winter over YRD, especially in January and February, is as high as in March to May. The mean extinction Angström exponent was found higher than 1.20 throughout the year, indicating that small particles were predominant in the region, which is different from North China where coarse dust particles are found in abundance in spring due to recurrent dust storms. An obvious wavelength dependence in SSA, that is strong absorption at infrared wavelengths, was found in July and August, and this was due to biomass burning or industrial emissions. The inter-site differences in the absorbing aerosol optical properties have been discussed in the revised manuscript, and the analysis indicated a degree of spatial heterogeneity in the distributions of absorbing aerosols even though the AODs were relatively similar. The difference in aerosols was attributed to the complex emission sources that impact the urban

and rural sites in the YRD.

(3) The direct DARF-BOA values under clear conditions were also calculated, and the results showed strongly negative DARF-BOA in June, followed by March and September due to high aerosol extinction. The monthly DARF-TOA means under clear conditions varied smoothly (averaging -40 W/m^2) during two periods: one in January to May and the other October to December. The mean DARF-TOA values under clear conditions were about −20 W/m^2 at seven sites in July/August with obviously lower AOD and SSA over YRD region. In contrast, the DARF-TOA values under clear conditions at Shenyang (Northeastern China urban area), Beijing (Northern China urban area), and Xianghe (Northern China rural area) showed the negative peak during June to August due to the large aerosol extinctions in summer season. The calculations also showed that the DARF-TOA was positive from April to October when the $SSA_{440\ nm}$ was < 0.80, and there was strong wavelength dependence, probably as a result of the burning of crop residues.

(4) As for the aerosol type classifications, we used the SSA, FMF, and EAE values to classify the absorbing weakly-absorbing particles in the fine and coarse modes. The results showed that the aerosol absorption is weak to moderate in the YRD region, and the fine-mode particles has an especially large contribution to the high percentage of absorbing particles at Hangzhou.

3. Details are still lacking in how RT simulations are conducted (aerosol layer altitude, cloud conditions, RF definition, etc), and the RT code has too large error/uncertainty (L180).

Response: Following to the reviewer's suggestion, more information has been added to the paper to explain how the RT simulations are done; the paper now includes a definition of RF, and it explain the assumptions regarding the altitude of aerosol layer , gas absorption, etc. We apologize for inaccurate earlier descriptions that were confusing to the reviewer. In this study, the authors investigated the direct aerosol radiative forcing (DARF) under free cloud conditions because the calculations were made based on the aerosol microphysical and optical parameters retrieved from ground-based measurements under free cloud conditions Details concerning the DARF calculations and error/uncertainty descriptions are highlighted (in RED) in Section 2 of the revised manuscript. Furthermore, the description of RT code error/uncertainty has been corrected as following in the present revision as follows:

"The flux calculations were performed for a multi-layered atmosphere with the US standard 1976 atmosphere model for gaseous distributions and single fixed aerosol vertical distribution (exponential with an aerosol height of 1 km) (Gacia et al., 2008). As these authors have pointed out, solar fluxes calculated using the module described above show excellent agreement with ground-based measurements of solar radiation (slope of 0.98 ± 0.00 and bias of -5.32 ± 1.00 W/m2) with a correlation of 99%. There is a small overestimation of +9 ± 12 Wm-2 of the observed solar radiation at the surface in global terms, and this corresponds to a relative error of +2.1 ± 3.0%. The differences range from +14 ± 10 Wm-2 to +6 ± 13 Wm-2 for urban-industrial and biomass burning aerosols, respectively. The errors are expected to be of the same magnitude at the TOA, since the same methodology and inputs are used at both levels (gaseous and aerosol distribution, radiative model, etc)."

4. I encourage the authors to formulate the most interesting and important findings from their data, rather than indulge themselves in the plain report of everything as what it is. It seems to me, the most important finding is that from all aspects, the seven sites show very little differences and indicate the presence of very similar aerosol and climate conditions.

Response: The authors agree with the reviewer's suggestions. In previous version, we simply presented the data without an in-depth analysis of the results. In the revised version, the most interesting and important findings from our data analysis have been highlighted and discussed in detail (Please see Response 2 above and also the revised manuscript).

Following up on the reviewer's important comments, we initially consider the data for the seven sites to investigate climatology of aerosol microphysical and optical characteristics over the YRD region. The seven sampling sites in this study are located in an area of 170 x 40km, and they include one densely populated urban site (Hangzhou), five urban center sites in smaller cities (LinAn, Fuyang, Jiande, Xiaoshan and Tonglu), and one rural site (ChunAn). Although there were generally small differences among sites, the results showing very similar aerosol and climate conditions provide a broadly representative picture of the aerosol populations over the whole YRD region. This is particularly true given that most of the previous studies were conducted at a single site and not for an extended time. Furthermore, even though the aerosols were relatively similar among seven sites, there are also some subtle differences in the aerosol optical properties, DARF, and aerosol type classification between the urban and rural areas. These differences reflect the impacts of local anthropogenic effects over specific parts of the YRD.

5. Comparison to MODIS should not be a goal of this study, as pointed out in the first round review, and should be reformulated/reduced.

Response: According to the first round review suggestion, the comparison to MODIS retrievals has been deleted but likely will be discussed in future work.

6. English needs improvement in quite a few places (misspells, typos, grammar, etc), a sign of haste and carelessness.

Response: The English has been polished by a native speaker carefully.

Reviewer 3

I found authors have addressed most of the reviewers's comments to improve the quality of the article. I only have a few minor and technical comments for the authors to consider.

Response: Thanks for the reviewer's help to improve the quality. We added "The authors would like to thank the three anonymous reviewers and the editor for their constructive suggestions and comments in the revised manuscript.

1. In terms of the paper structure, I still think it is not appropriate to present the validation of MODIS AOD in section 3.2. This section presents the aerosol optical properties from Sun Photometers, and satellite of validation belongs to one of the applications of the CARSNET products. So, please consider to use a standalone section for MODIS validation.

Response: Following both reviewers' suggestions, the comparison to MODIS retrievals has been deleted in the revised paper.

2. Line 39-40: "volume fraction" means the fraction of volume, which is not appropriate to be used as volume. I recommend to replace it with "modal volume" or "fractional volume". And the units of volume here should um^3/um^2, volume of aerosol particle per um^2 area. This correction should be applied across the entire article.

Response: The abstract has been rewritten according to the reviewers' suggestions. In the revised abstract and elsewhere, the term of "volume fraction" has been represented as "volume" or "fractional volume". The units of volume in the revised manuscript have been corrected as "$\mu m^3/\mu m^2$" across the entire paper.

3. Line 42: 440nm -> 440 nm. This also applies elsewhere in the article.

Response: The term of "440nm" has been correct into "440 nm" throughout the revised manuscript.

4. Line 48: 10km -> 10 km; 3km -> 3 km. Again, elsewhere in the article.

Response: Because the part about the comparison to MODIS retrievals has been deleted, the terms of "10km" and "3km" have been removed in the revised paper.

5. Line 54-57: First, change "radiative forcing" into "direct radiative forcing", because indirect radiative forcing was not considered. Second, it lacks innovation to simply say "aerosols

causes negative forcing ...", because negative radiative effect of aerosol over low-reflectivity surface has been well known. So, present the numbers. Third, it reads "… the lower surface albedo in a unique geographical climate condition of better vegetation in the YRD region than in north/northeast China." So, does it means that aerosols in north/northeast China exert a positive radiative effects? Please make it clear.

Response: According to the reviewer's suggestion (1) the term of "radiative forcing" has changed into "direct radiative forcing"; (2) the detailed aerosol direct radiative forcing numbers have been added in the revised paper; (3) to avoid confusion to readers, this sentence in question was removed from the revised paper.

6. Figure 1: I don't see any description of the two red river lines on the map. Description maybe needed in the figure caption, otherwise, consider to remove them.

Response: According to the reviewer's suggestion, the two red river lines representing the Yellow River and Yangtze River on the map in Figure 1 have been removed in the revised paper.

7. Lines 222, 406, 464, and 557: Redundant section titles. Consider to remove "Aerosol optical properties of"

Response: The section titles have been shortened in the revised paper.