

We appreciate the reviewer's valuable comments and constructive suggestions which help us improve the quality of the manuscript. We have carefully revised the manuscript according to these comments. Point-to-point responses are provided below. The reviewer's comments are in black, our responses are in blue and the corresponding changes in manuscript are in red.

Reviewer #1

The present study is highly important for both SKYNET products and for other sunphotometric instruments and networks that could potentially benefit from the enhanced approach of SKYRAD inversions methodology. A detailed comparison between the two versions of SKYRAD is missing from the literature and it is always a question for scientists handling SKYNET data. Stations selected for the study seem to provide a sufficient amount of data for this comparison. Additionally, authors have exploited these datasets to provide climatology of aerosol properties at both measuring locations. However, the manuscript lacks of explanations on the causes of differences between the v4.2 and v5.0 retrievals and sufficient evidence on the actual seasonal variability of aerosols in the two regions. Algorithms of both versions are clearly described, but it is crucial to pinpoint and discuss the way the differences between the versions affect the retrievals. Since the two algorithms are not treated as a "black box", it should be more clear which physical processes affects the retrievals and which atmospheric conditions could lead to highest uncertainty. At least some discussion on the uncertainty of each variable in each approach should be provided. Also, the part about the seasonal variability of aerosols properties, results are presented but not investigated and discussed in the level expected for a scientific study. Majority of readers are not unfamiliar with local weather systems and patterns, emissions, and these should be described in the manuscript. Thus, I suggest that the manuscript should be considered for publication in AMT after a major revision addressing these concerns.

Response: Thank you for your valuable comments and constructive suggestions. We have carried out additional experiments and found that the calibration constants in the previous experiments were incorrect, so we corrected them and re-carried out the experiments and numerical tests; some results and figures have been updated and represented in the following response.

We have added more details related to the graphical, climate and major chemical compositions in PM_{2.5} in the two sites. Some of the new figures and comments about inter-comparisons results

between V5.0 and V4.2 have been shown in the following comments. Meanwhile, we have also investigated some parameters linked to the SSA differences between the V5.0 and V4.2, the seasonal variation of aerosol have been discussed combining the possible emission sources and prevailing wind based on more data and references as shown in the following comments.

Specific comments

P2 15-7 This should be divided in two sentences because it is confusing.

Response: The sentence has been divided into two sentences in the revised manuscript.

P3 117 I assume this precision is for the sky radiance measurement, but it should restated to be clear.

Response: This precision is for the sky radiance measurement. To avoid confusion, we have added the following revised the sentence to ‘The typical measurement interval of the sky radiance is 10 min’ in the revised manuscript.

P3 118 Some details on the calibration of these instruments should be added.

Response: Following the reviewer’s suggestion, we have added the following comments in the revised manuscript: Improved Langley (IL) plot method is used in this study to determine the temporal and spectral calibration constants for direct intensity (F0) with accuracy of about 1.0–2.5 %, depending on the wavelength (Nakajima et al., 1996; Campanelli et al., 2004). The calibration by IL plot method is made daily, the variation of F0 due to instrumental drift can be quickly spotted, and then appropriate corrections to data can be applied exactly from the period in which the deviation occurred (Campanelli et al., 2004).

P3 119-24 More detailed description of the locations is needed.

Response: We have added a map in the revised manuscript to show the locations of the two SKYNET sites in this study as shown in the following figure.

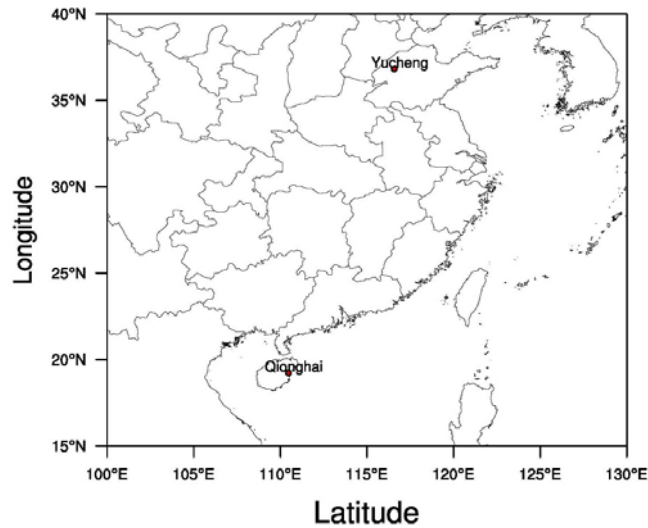


Figure 1: The locations of the two SKYNET sites in the study

In section 2.2 Site description, we have added some more details including monsoon, temperature, and precipitation. We have added the following descriptions in the revised manuscript.

The Qionghai site of SKYNET (19.23°N, 110.46°E, 24 m a.s.l.), which was located in the eastern part of Hainan Island, was mainly influenced by East Asia monsoons and typhoons. During summer, the dominant wind is from south to southeast, summer monsoon from the South China Sea and West Pacific brought most of the annual rainfall to the island (Zhu et al., 2005), whereas the winter monsoon from Inner Mongolia carries dry winds to the area (Zhu et al., 2005; Peel et al., 2007; Yin et al., 2002). Annual average rainfall in Qionghai is estimated about 1653.4 mm. Maximum high temperature occurs in July, with monthly average of 28.6 °C, monthly lowest temperature occurs in January, with monthly average of 19.1 °C (Yin et al., 2002).

The other measurement site in this study was located in rural Yucheng (36.82°N, 116.57°E, 22 m a.s.l.), Shandong Province, China, which is almost in the centre of the North China Plain. The selected site is in an open field surrounded by farmland. The region belongs to semi-humid and temperate monsoon climate zone, characterized by a mean annual temperature of 21 °C and mean annual precipitation of 610 mm mainly distributed in summer months (Chen et al., 2012). Yucheng and the surrounding areas are famous for their agriculture (e.g., wheat and corn) and grazing land (e.g., donkeys and chickens). In addition, the site near 20 to 30 km radius located several factories in the production of inorganic and organic fertilizers (Wen et al., 2015), and the application of fertilisers to farmland emitted a great deal of NH₃ (Zhao et al., 2012). Meanwhile, Yucheng was located in the downwind of the Beijing-Tianjin-Hebei region, long-distance transport of sources of industrial

pollution and biomass burning contributed significantly to the concentrations of pollutants in Yucheng (Lu et al., 2016).

In addition, based on the results simulated by the Community Multi-scale Air Quality model with the 2D Volatility Basis Set (CMAQ/2D-VBS) (23), we have added the following comments and figure in the revised manuscript to describe the major chemical compositions in PM_{2.5} and their percentage contribution to PM_{2.5} at the two sites.

It is well known that OC, EC, SO₄²⁻, NO₃⁻ and NH₄⁺ were the dominant chemical components in PM_{2.5} (Tao et al., 2017). The above-mentioned five major components over the two sites were discussed below based on the results simulated by the Community Multi-scale Air Quality model with the 2D Volatility Basis Set (CMAQ/2D-VBS) (23) at 36- × 36-km resolution with emission inputs derived from a Chinese emission inventory developed and updated to 2015 with details in these studies (Wang et al., 2014; Zhao et al., 2018). The contributors to carbonaceous aerosols in China mainly include coal combustion, vehicle exhaust and biomass burning, etc (Liu et al., 2018). As shown in Fig. 9, the concentrations of OC were significantly higher than that of EC at Qionghai, likely due to the mixed contributions of atmospheric chemical reactions and primary anthropogenic sources to OC (Cao et al., 2004). The nitrate accounted for a large fraction of PM_{2.5} in Yucheng, it was strongly related to the high emission levels of NH₃ and O₃ in Yucheng (Wen et al., 2015).

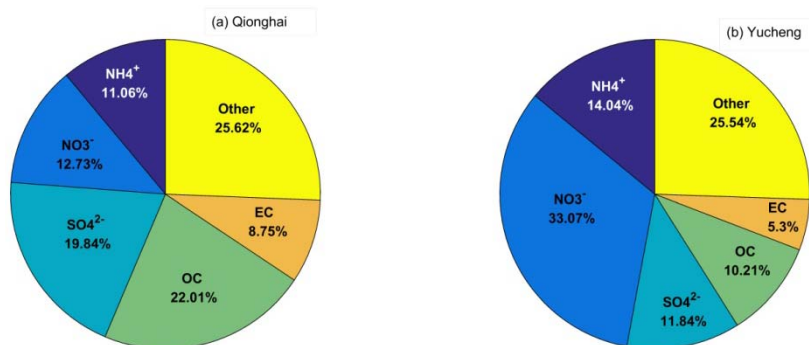


Figure 9: Percentage (%) contribution of NO₃⁻, SO₄²⁻, NH₄⁺, OC and EC to PM_{2.5} mass in Qionghai (a) and Yucheng (b) in 2015

P5 I10 More details on the quality control and cloud screening procedures should be provided.

Response: The standard process of quality control in Skyrad.pack V4.2 and V5.0 applies a retrieval error between observations and calculated theoretical values by using retrieval values, σ_{obs}

$$\sigma_{\text{obs}} = \sqrt{W_e \sum_i \left(\frac{\tau_{\lambda_i}}{\tau_{\lambda_i}^{\text{meas}}} - 1 \right)^2 + W_p \sum_i \sum_j \left[\frac{R_{\lambda_i}(\theta_j)}{R_{\lambda_i}^{\text{meas}}(\theta_j)} - 1 \right]^2}$$

where $(\tau_{\lambda_i}^{\text{meas}}$ and $R_{\lambda_i}^{\text{meas}})$ and $(\tau_{\lambda_i}$ and $R_{\lambda_i})$ are measured and retrieved observation vectors for the AOD and relative sky radiance, N_i , N_j , and $N_{\text{total}} = N_i + N_i \times N_j$ indicate the number of measured wavelengths, scattering angles, and their total, respectively, $W_e = W_p = 1/N_{\text{total}}$. In V4.2, the data if the value of σ_{obs} is larger than 0.2, but σ_{obs} is set 0.07 as a threshold for data rejection in V5.0. There are some other differences between V4.2 and V5.0 on the issue of quality control of observation data and cloud screening (Hashimoto et al., 2012).

We have added the above comments in the revised manuscript.

P5 117. It would be useful to report the number of measurements fulfilling this criterion at both sites

Response: Thank you for your kind comments. In the new experiment, we have added the measurement data from March to December in 2015. There are 3995 measurements for 436 days fulfilling V4.2 criterion and 2159 measurements for 355 days fulfilling V5.0 criterion over Qionghai. There are 13061 measurements for 577 days fulfilling V4.2 criterion and 7921 measurements for 473 days fulfilling V5.0 criterion over Yucheng.

The inter-comparisons of aerosol properties between V5.0 and V4.2 were based on 1397 measurements and 5830 measurements over Qionghai and Yucheng, respectively.

We have added the above comments in the revised manuscript.

P5 127 This sentence indicates that v 5.0 is more erroneous in coarse mode. Is there more evidence on that? Is that strictly due to algorithmic reasons? More discussion is needed on this effect.

Response: In the case of a large amount of coarse particles of the dust-like aerosol type with radius greater than 10 μm existing, the numerical tests performed by Hashimoto et al (Hashimoto et al., 2012) showed that V4.2 could retrieve the SDF relatively well, including the coarse mode, in comparison with V5.0, because the smoothness condition given by Eq. (2) in the manuscript allowed the retrieved SDF to be distributed beyond 10 μm radius, on the other hand, V5.0 underestimated the coarse mode of the SDF because of the strong SDF constraint condition given

by Eq. (5) with a small model radius $r_{m2} = 2.0 \mu\text{m}$ for the coarse mode SDF (Hashimoto et al., 2012).

In the case of cirrus contamination existing, the sensitivity tests results shows that V4.2 retrieved the aerosol size distribution function (SDF) including contaminating cirrus particles larger than $10 \mu\text{m}$, but version 5 successfully filtered out the cirrus particles by the constraint of a reduced SDF for particles with radius greater than $10 \mu\text{m}$ (Hashimoto et al., 2012).

We have added the above discussion and figure in the revised manuscript. We have also removed the sentence in P5 127 that indicates that v 5.0 is more erroneous in coarse mode.

Figure 1. It is really difficult to visually distinguish the differences between the two versions for most bins. Probably a different approach should be also demonstrated here (absolute differences, relative differences? histogram?) to facilitate reader's comprehension. Also a x axes label is missing.

Response: We have replaced Fig.1 with the following figure which shows the retrieved monthly volume size distribution between SKYRAD V4.2 (red lines) and V5.0 (blue lines) for Qionghai (dotted line) and Yucheng (solid lines) during February 2013 to December 2015. As shown in the following figure, V4.2 showed a tri-model pattern with three peak volume at radius of 0.25, 1.16, 11.31 and 0.25, 1.69, 11.31 in July and September over Yucheng, respectively. Figure 3 also showed that there were larger differences in volume SDF of the coarse mode between V4.2 and V5.0 at Qionghai than those at Yucheng in most months.

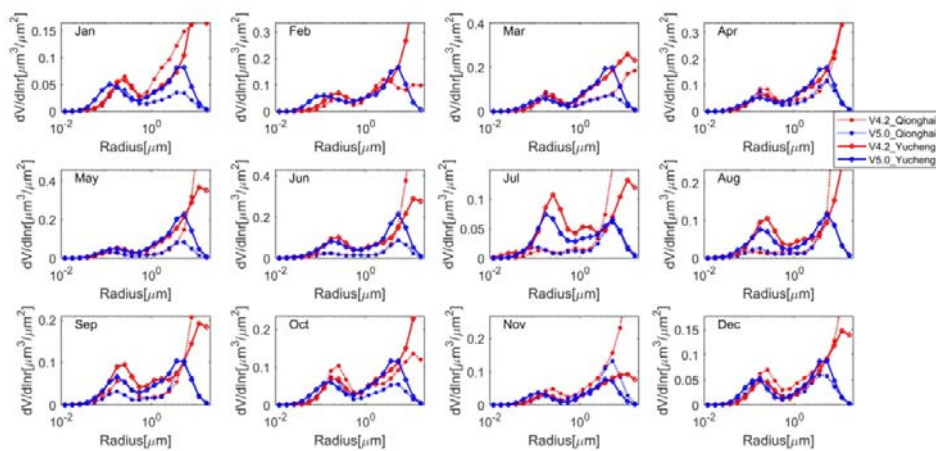


Figure 3: Retrieved monthly volume size distribution between SKYRAD V4.2 (red lines) and V5.0 (blue lines) for Qionghai (dotted line) and Yucheng (solid lines) during February 2013 to December 2015

We have added the above figure and comments in the revised manuscript.

Paragraph 3.2 Some physical interpretation and discussion about these differences is missing. Are they explained strictly algorithmically or is there some natural process driving them? Differences of SSA are really high, and have opposite behavior (more absorbing for v 5.0 in Qionghai and more scattering in Yucheng). Keep in mind that SSA values in the atmosphere have a very small range, and these differences are very high. SSA at 0.92 and 0.86 (example at Qionghai at January) indicate totally different types of aerosol. In addition, the different behavior at the two sites, makes it difficult to assume some systematical bias. Since there are no other independent data to validate which version is closest to the actual condition, I strongly suggest to investigate further this behavior. In the scientific literature you could find a number of approaches to select depending on the data available, but it is crucial at this point to have some evidence on the validity of the retrievals.

Response: We found that the calibration constants were incorrect in the previous experiment, so we corrected them and re-carried out the experiments, some new results and conclusions have been got. The following figure presents the compared results of SSA between SKYRAD V4.2 and V5.0 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai and Yucheng during February 2013 to December 2015. SSAs by V5.0 correlated to SSAs by V4.2 with $R= 0.88, 0.87, 0.90, 0.88$ and 0.92 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai , respectively. The SSA values computed from V5.0 had correlation coefficients around $0.95, 0.95, 0.96, 0.94, 0.91$ at wavelengths of 400, 500, 670, 870, and 1020 nm over Yucheng.

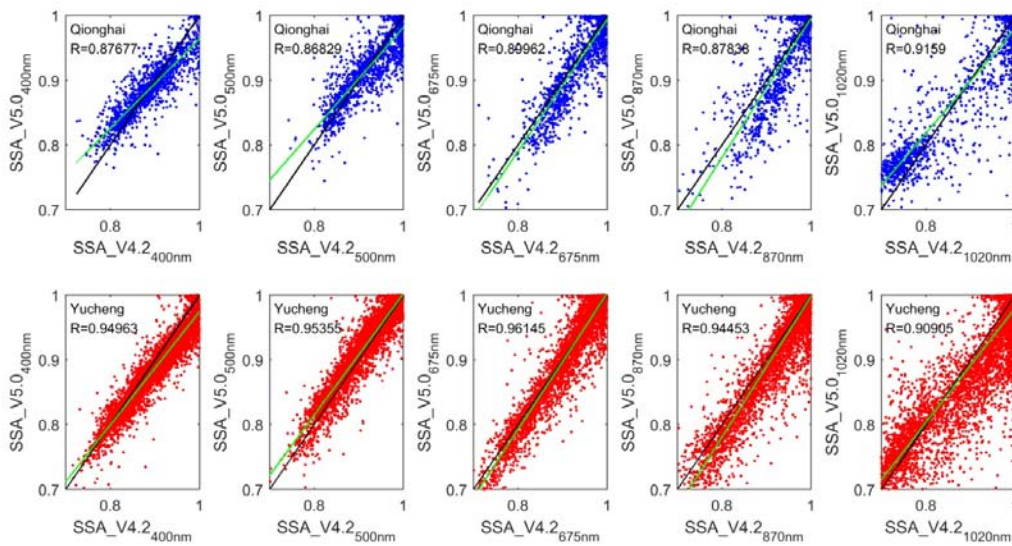


Figure 4: Scattergrams of the single scattering albedo between SKYRAD 4.2 and 5.0 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai and Yucheng during February 2013 to December 2015. Only data with $AOD_{500nm} > 0.2$ are shown. The green line means the fitted linear regression curve.

We have added the above comments in the revised manuscript.

Figure 2. bar plots for mean monthly values and showing only the higher part of the error bar (which I assume is standard deviation but nowhere stated) is confusing. I suggest to visualize in another way.

Response: We have replaced Fig.2 with the scattergrams of the single scattering albedo between V4.2 and V5.0 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai and Yucheng during February 2013 to December 2015 as shown in the above.

P7 I17 Frequency distributions are plotted in figure3. Where probability distributions mentioned here could be found?

Response: We have replaced Fig.3 with scattergrams of the imaginary and real part of the complex refractive index (m_i) results between V.2 and V5.0 as bellow. As shown in Fig. 5, the m_i values by V5.0 were linearly correlated with m_i by V4.2 with $R=0.8947, 0.8661, 0.8658, 0.8370, 0.9131$ at wavelengths of 400, 500, 675, 870 and 1020 nm in Qionghai. The correlation coefficients between m_i by V5.0 and those by V4.2 at the five wavelengths were all higher than 0.89 over Yucheng.

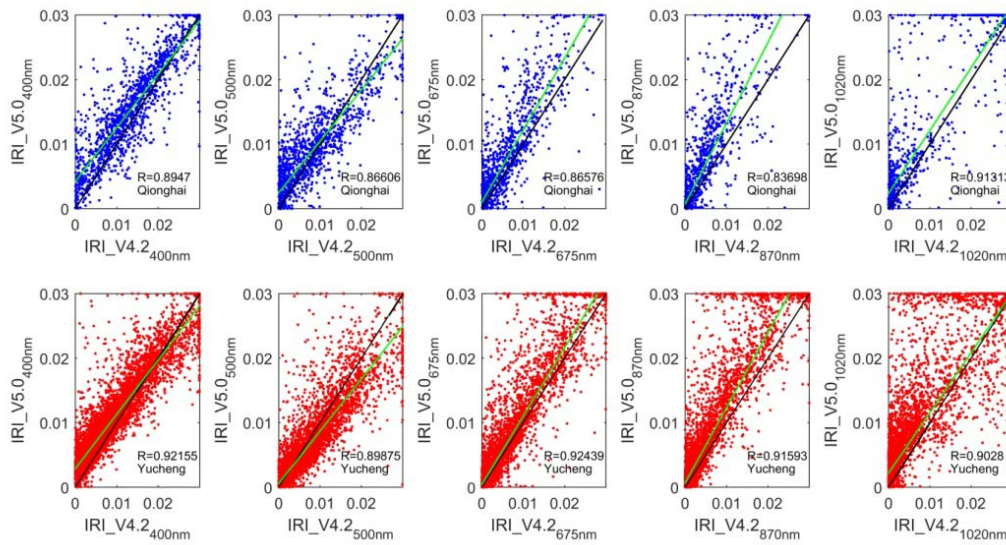


Figure 5: Scattergrams of the imaginary part of the complex refractive index (m_i) results between SKYRAD 4.2 and 5.0 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai and Yucheng during February 2013 to December 2015. The green line means the fitted linear regression curve.

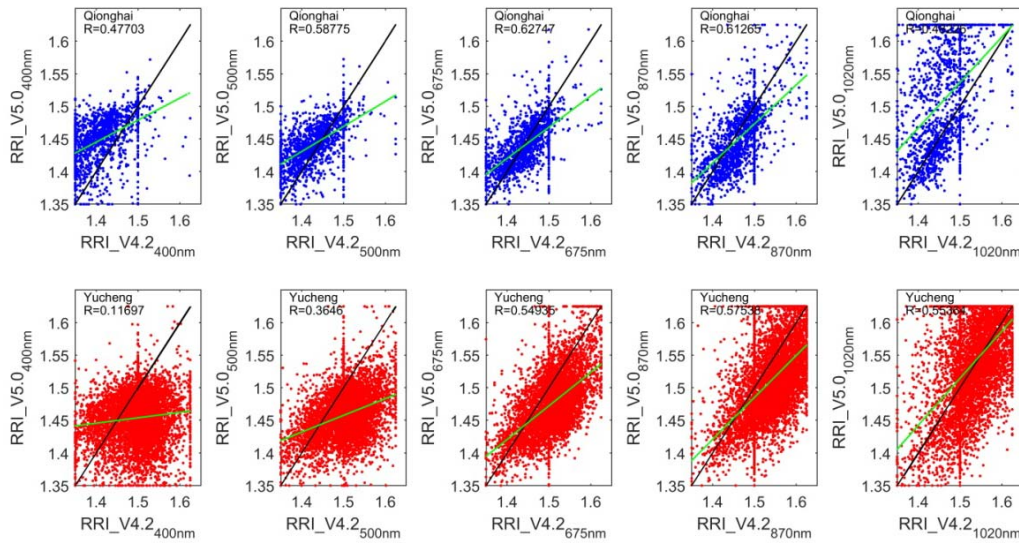


Figure 6: Scattergrams of the real part of the complex refractive index (m_r) results between SKYRAD 4.2 and 5.0 at wavelengths of 400, 500, 670, 870, and 1020 nm over Qionghai and Yucheng during February 2013 to December 2015. The green line means the fitted linear regression curve.

Figure 3. x axis label is missing Figure 4. x axis label is missing

Response: We have replaced Fig.3 and Fig.4 with the above figures.

General comment for 3.2-3.3. First, a more uniform approach on the presentation of results should be applied. Treating histograms for refractive index and monthly averages for SSA, makes the datasets incomprehensible, since cannot be easily combined and provide a conclusion on the behavior. Also, some conclusions should be reached linked to the differences of the two versions and the causes of the variations. For that purpose, there should be some discussion about the algorithmic differences and the outputs. Finally, it is important to understand whether other parameters are linked to the differences. At least it should be investigated the corresponding aerosol loads(AOD) for each case. Does the difference increase/decrease with higher AOD? Is the elevation of the sky radiance measurements linked to the differences between the two versions?

Response: Based on the new experiment results, the inter-comparison results of SSA and refractive index have been all presented in the form of scatter grams as above. SSA and the imaginary part of the complex refractive index (m_i) from V5.0 both had higher correlation coefficients with those from V4.2 in Yucheng than in Qionghai.

V4.2 uses the iterative relaxation method of Nakajima et al. (1983, 1996) to derive the aerosol size distribution function (SDF) and other parameters, the retrieved refractive index can only be chosen from the predefined set of values. V5.0 uses the non-linear maximum likelihood method defined by Rodgers (2000) which has a strong dependence on the estimation of the first-guess solution. V5.0 used an a priori SDF of a bimodal log-normal function, we have compared the differences between retrieved SSAs at 500 nm by V5.0 and V4.2 when set $rm_2 = 1.5, 1.8, 2.0(\text{default}), 2.5$ and 3.0 in Skyrad.pack V5.0 based on the measurements in 2014. As shown in the following figure, SSAs by V5.0 correlated to SSAs by V4.2 with $R = 0.860, 0.837, 0.855, 0.809$ and 0.226 when $rm_2 = 1.5, 1.8, 2.5$ and 3.0 in V5.0 over Qionghai, respectively. The correlation coefficient between SSA by V5.0 and V4.2 was the highest while setting rm_2 as 2.0 (the default value) at both the two sites.

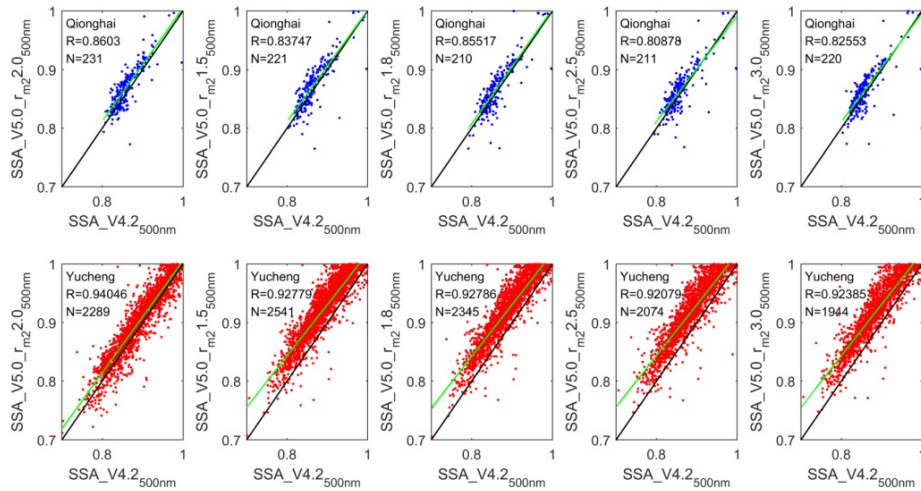


Figure 7: Scattergrams of retrieved SSA between SKYRAD V4.2 and V5.0 when $r_{m2}=2.0$ (default), 1.5, 1.8, 2.5 and 3.0 for Qionghai (a) and Yucheng (b) in 2014. r_{m2} represents the model radius for the coarse mode SDF. The green line means the fitted linear regression curve.

We have added the above figure and comments in the revised manuscript.

Following the reviewer's suggestion, we have investigated whether the total amount of aerosols in the atmosphere were linked to the difference in SSA between the two versions. As shown in the following figure, the SSA differences between the two versions decreased with the corresponding AODs increased at both the two sites.

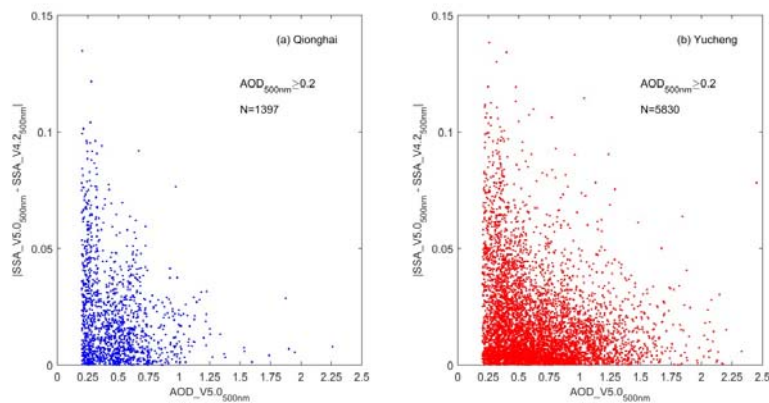


Figure 8: Scattergrams of the SSA differences at 500nm between V5.0 and V4.2 (defined as: $|SSA_{V5.0,500nm} - SSA_{V4.2,500nm}|$) and the corresponding AODs at wavelengths of 500 nm by V5.0 during February 2013 to December 2015.

We have added the above figure and discussions in the revised manuscript.

We have also investigated the variation of the SSA differences between the two versions with solar height over the two sites. As shown in the following figure, the solar height isn't likely linked to

the differences between the two versions.

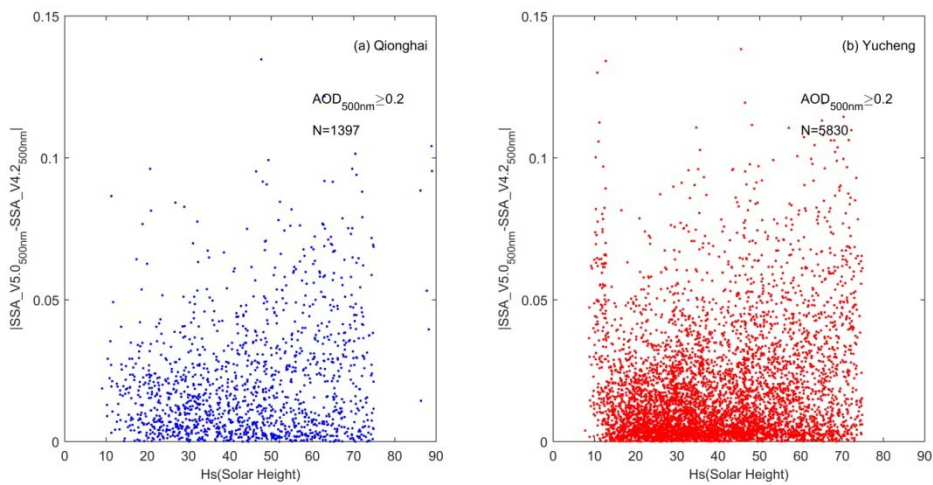


Figure: Scattergrams of the SSA differences at 500nm between V5.0 and V4.2 (defined as: $|\text{SSA}_{V5.0_{500\text{nm}}} - \text{SSA}_{V4.2_{500\text{nm}}}|$) and the corresponding solar height over Qionghai (a) and Yucheng (b).

P9 116-17 Some reference or some data are needed to provide evidence about the meteorological argument.

Response: We have added more meteorological description of the two sites and the corresponding references as shown in the above response to the comment ' P3 119-24 More detailed description of the locations is needed' .

P9 120 Further discussion and evidence are needed to support this argument.

Response: We have added the following comments in the revised manuscript.

The prevailing winds in Yucheng were from the northwest in winter and spring. Yucheng was in the downwind of Hebei province where located many industrial enterprises emitted pollutants including secondary inorganic aerosols (Tao et al., 2017; Zhao et al., 2018c). The high AOD in spring was likely related to the presence of the dust particles transported from the northwest of China and secondary inorganic aerosols emitted from enterprises in Hebei (Tao et al., 2017).

P10 114. By definition, SSA will decrease when absorbing aerosols increase. This sentence does not provide any explanation on the behavior. More detailed discussion should be added on these results.

Response: We have added the following comments about SSA seasonal variation in the revised manuscript.

The lowest seasonal average SSA was observed in winter, which was probably attributable to the regional transport of the air masses originated from the regions outside of Hainan province in Eastern China, where a great amount of coal was used for industrial enterprises and emitted high concentrations of OC and EC (Liu et al., 2018). In Yucheng, the seasonal pattern of SSA was consistent with AOD, the lowest seasonal average SSAs were also observed in winter due to carbonaceous aerosols increasing by heating activities and biomass burning, seasonal average contributions of carbonaceous aerosols were evidently higher in cold seasons than in warm seasons (Tao et al., 2017). High concentrations of fine particulate nitrate were frequently observed in summer in Yucheng (Wen et al., 2015), likely to cause the high SSA in summer.

Paragraph 3.4.2 Since for SSA the selection of version 4.2 or 5.0 could lead to different conclusions on the type of aerosols, some discussion on that issue should be added here. P10 l26-17 Since the algorithm uses bimodal fits, there was no way to find a different distribution

Response: Based on the new experiment results, the SSA and m_i had relatively high correlation coefficients between V4.2 and V5.0 with default rm_2 values. In addition, some tests by Hashimoto et al showed that the SDF setting in V5.0 was useful for detecting ill-conditioned data caused by cirrus contaminations, horizontally and/or temporally inhomogeneous aerosol stratification, and so on (Hashimoto et al., 2012). So we still chose the retrieved results by V5.0 to analyze the seasonal variability of the aerosol optical properties over Qionghai and Yucheng.

P11 l2-4. Why anthropogenic aerosols should decrease in winter/spring? Are there any information on the human activities in the area? Why sea salt aerosols increase? Also, some information about the monsoonal influence in the region should be added for the readers that are unfamiliar with local climatology (preferably at the site description section at page 3)

Response: The new seasonal averaged volumes of the different aerosol size distributions ($dv/d\ln r$) in Qionghai and Yucheng are shown as follows. In Qionghai, the fraction of the fine aerosol particles was much smaller in summer than for the other seasons, the summer meteorological conditions such as high wind speeds, high mixing heights, and the fresh air masses originated

from or passed through the sea, which may be contributable to the decrease of pollutant concentrations (Liu et al., 2018) and introducing some sea salt particles of a relatively large size. As shown in the following figure, the seasonal averaged peak of fine mode and coarse mode were both in winter, the air masses of transport were mainly originated from the mainland China, fine and coarse particle were both long range transported to Qionghai in winter (Wu et al., 2011; Liu et al., 2018).

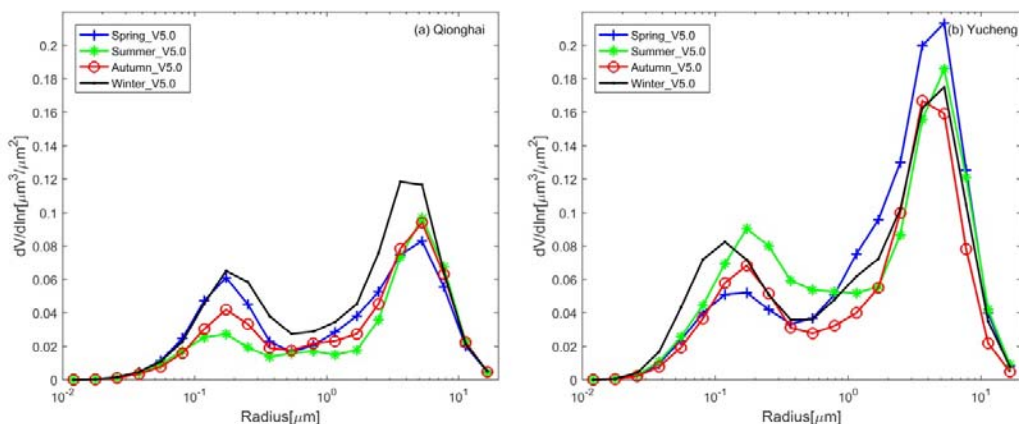


Figure 11: Seasonally averaged volumes of the different aerosol particle size distributions based on SKYRAD V5.0 over Qionghai (a) and Yucheng (b) for the period from February 2013 to December 2015.

We have added the above figure and discussion in the revised manuscript.

P11 16-8 Also fine mode is very high in summer (compared to Qionghai). Any interpretation on that? The only source of large particles in the area is dust long transport or are there any other sources?

Response: High concentrations of fine particulate nitrate were frequently observed in summer in Yucheng (Wen et al., 2015), likely to cause the high SSA in summer. The prevailing winds in Yucheng were from the northwest in winter and spring, Yucheng was in the downwind of Hebei province where located many industrial enterprises emitted pollutants including secondary inorganic aerosols (Tao et al., 2017; Zhao et al., 2018c). The aerosol was not only from winter heating but also from regional transport, the fine-mode and coarse-mode particles was both high in winter in Yucheng. The volume of the coarse aerosol particles relative to the whole was much larger than for the other seasons in spring in Yucheng probably because of the presence of the dust particles transported from the northwest of China and secondary inorganic aerosols emitted from

enterprises in Hebei (Tao et al., 2017).

We have added the above discussion in the revised manuscript.

P11 18. Also fine mode seemed to peak to almost double values in summer/winter compared to spring/autumn. Is there any explanation for this behavior?

Response: Yucheng and the surrounding areas are famous for their agriculture (e.g., wheat and corn) and grazing land (e.g., donkeys and chickens). In addition, the site near 20 to 30 km radius located several factories in the production of inorganic and organic fertilizers (Wen et al., 2015), and the application of fertilisers to farmland emitted a great deal of NH₃ (Zhao et al., 2012). High concentrations of fine particulate nitrate were frequently observed in summer in Yucheng (Wen et al., 2015), likely to cause the high SSA in summer.

Figure 6. X axes label is missing

Response: In all the new figures, we have checked X axes label and other details.

General comment for 3.4 I suggest to summarize the types and variations of aerosols in both sites in a more descriptive way at the end. Also, It would be very useful a discussion –based on earlier paragraphs- on the properties and conditions that both version come together and the conclusions that have higher uncertainties due to the deviations between the algorithms.

Response: Based on the new experiment results, the SSA and m_i had relatively high correlation coefficients between V4.2 and V5.0. Quality control and cloud screening procedures in V5.0 are stricter than V4.2 (Hashimoto et al., 2012). So we still chose the retrieved results by V5.0 to analyze the seasonal variability of the aerosol optical properties over Qionghai and Yucheng.

The AOD showed high values in spring, autumn and winter but decreased to minimum in summer over Qionghai likely related to summer monsoon from the South China Sea and West Pacific that brought most of the annual rainfall to the island, whereas the winter monsoon from Inner Mongolia carried the air masses from the mainland China to Qionghai. In Yucheng, the maximum seasonal averaged AOD and SSA both appeared in summer due to the hygroscopic effects of secondary inorganic aerosols. The fraction of the fine aerosol particles over Qionghai was much smaller in summer probably related to wet deposition, more precipitation in the summer can lead

to more efficient removal of aerosol. Aerosol was not only from winter heating but also from regional transport, the fine-mode and coarse-mode particles was both high in Yucheng in winter. The volume of the coarse aerosol particles relative to the whole was much larger than for the other seasons in spring, probably due to the presence of the dust particles transported from the northwest of China and pollutants emitted from enterprises in Hebei.

P12 116-18 There is no evidence in the study of the cause of this behavior (algorithm or type of aerosols?). More work should be done before coming to this conclusion.

Response: We have replaced this conclusion with the following conclusion based on the above analysis. The location and distribution of major industrial sources, intensity of local minor sources such as winter heating, and prevailing wind directions together caused the slightly different magnitudes of seasonal variations among the two sites discussed above.

References

- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A.J., Ramanathan, V., and Welton E.J.: Reduction of tropical cloudiness by soot, *Science*, 288, 1042–1047, doi: 10.1126/science.288.5468.1042, 2000.
- Bi, J. R., Shi, J. S., Xie, Y. K., and Liu Y. Z.: Dust Aerosol Characteristics and Shortwave Radiative Impact at a Gobi Desert of Northwest China during the Spring of 2012, *J. Meteor. Soc. Japan*, 92A, 33–56, doi: 10.2151/jmsj.2014-A03, 2014.
- Cai J. X., Guan, Z. Y., and Ma, F. H.: Possible combined influences of absorbing aerosols and anomalous atmospheric circulation on summertime diurnal temperature range variation over the middle and lower reaches of the Yangtze River, *J. Meteor. Res.*, 30(6), 927–943, doi: 10.1007/s13351-016-6006-1, 2016.
- Campanelli, M., Nakajima, T., and Olivieri, B.: Determination of the solar calibration constant for a sun-sky radiometer: Proposal of an in-situ procedure, *Appl. Opt.*, 43, 651–659, doi:10.1364/AO.43.000651, 2004.
- Campanelli, M., Lupi, A., Nakajima, T., Malvestuto, V., Tomasi, C., and Estelles, V.: Summertime columnar content of atmospheric water vapor from ground-based Sun-sky radiometer measurements through a new in situ procedure, *J. Geophys. Res.*, 115, D19304, doi:10.1029/2009JD013211, 2010.

- Che, H., Shi, G., Uchiyama, A., Yamazaki, A., Chen, H., Goloub, P., and Zhang, X.: Intercomparison between aerosol optical properties by a PREDE skyradiometer and CIMEL sunphotometer over Beijing, China, *Atmos. Chem. Phys.*, 8, 3199–3214, doi:10.5194/acp-8-3199-2008, 2008.
- Che, H. Z., Xia, X. A., Zhu, J., Wang, H., Wang, Y. Q., Sun, J. Y., Zhang, X. C., Zhang, X. Y., and Shi, G. Y.: Aerosol optical properties under the condition of heavy haze over an urban site of Beijing, China, *Environ. Sci. Pollut. R.*, 22, 1043–1053, <https://doi.org/10.1007/s11356-014-3415-5>, 2014.
- Che, H. Z., Qi, B., Zhao, H. J., Xia, X. A., Eck, T. F., Goloub, P., Dubovik, O., Estelles, V., Cuevas-Agulló, E., Blarel, L., Wu, Y. F., Zhu, J., Du, R. G., Wang, Y. Q., Wang, H., Gui, K., Yu, J., Zheng, Y., Sun, T. Z., Chen, Q. L., Shi, G. Y., and Zhang, X. Y.: Aerosol optical properties and direct radiative forcing based on measurements from the China Aerosol Remote Sensing Network (CARSNET) in eastern China, *Atmos. Chem. Phys.*, 18, 405–425, doi:10.5194/acp-18-405-2018, 2018.
- Chen, Z., Lu, C. and Fan, L.: Farmland changes and the driving forces in Yucheng, North China Plain. *Journal of Geographical Sciences*, 22(3), 563-573, 2012.
- Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, 312(5778), 1375–1378, doi:10.1126/science.1125261, 2006.
- Estellés, V., Campanelli, M., Utrillas, M. P., Expósito, F., and Martínez-Lozano, J. A.: Comparison of AERONET and SKYRAD4.2 inversion products retrieved from a Cimel CE318 sunphotometer, *Atmos. Meas. Tech.*, 5, 569–579, <https://doi.org/10.5194/amt-5-569-2012>, 2012a.
- Estellés, V., Smyth, T. J., Campanelli, M.: Columnar aerosol properties in a Northeastern Atlantic site (Plymouth, United Kingdom) by means of ground based skyradiometer data during years 2000–2008, *Atmos. Environ.*, 61, 180–188, doi :10.1016/j.atmosenv.2012.07.024, 2012b.
- Hashimoto, M., Nakajima, T., Dubovik, O., Campanelli, M., Che, H., Khatri, P., Takamura, T., and Pandithurai, G.: Development of a new data-processing method for SKYNET sky radiometer observations, *Atmos. Meas. Tech.*, 5, 2723–2737, <https://doi.org/10.5194/amt-5-2723-2012>, 2012.
- He, L. Y., Hu, M., Zhang, Y. H., Huang, X. F., and Yao, T. T.: Fine particle emissions from on-road vehicles in the Zhujiang Tunnel, China, *Environmental Science & Technology*, 42, 4461–4466,

2008.

Hensen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, *J. Geophys. Res.*, **102**(D6), 6831–6864, doi:10.1029/96JD03436, 1997.

Higurashi, A., Nakajima, T., Holben, B., Smirnov, A., Frouin, R., and Chatenet, B.: A study of global aerosol optical climatology with two channel AVHRR remote sensing, *J. Climate*, **13**, 2011–2027, 2000.

Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D., and Rudich, Y.: The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean, *Proc. Natl. Acad. Sci. U.S.A.*, **102**, 11207–11212, doi: 10.1073/pnas.0505191102, 2005.

Kim, D. H., Sohn, B. J., Nakajima, T., Takamura, T., Takemura, T., Choi, B. C., and Yoon, S. C.: Aerosol optical properties over east Asia determined from ground-based sky radiation measurements, *J. Geophys. Res.*, **109**, D02209, doi:10.1029/2003JD003387, 2004.

Li, M., Xie, L., Zong, X., Zhang, S., Zhou, L., and Li, J.: The cruise observation of turbulent mixing in the upwelling region east of Hainan Island in the summer of 2012. *Acta Oceanologica Sinica*, **37**(9), 1-12, 2018.

Liu, B., Zhang, J., Wang, L., Liang, D., Cheng, Y., Wu, J., Bi, X., Feng, Y., Zhang, Y., and Yang, H.: Characteristics and sources of the fine carbonaceous aerosols in Haikou, China. *Atmospheric Research*, **199**, 103-112, 2018.

Lu, W., Yang, L., Chen, J., Wang, X., Li, H., Zhu, Y., Wen, L., Xu, C., Zhang, J., Zhu, T. and Wang, W.: Identification of concentrations and sources of PM 2.5-bound PAHs in North China during haze episodes in 2013. *Air Quality, Atmosphere & Health*, **9**(7), 823-833, 2016.

Meng, C. L., and Xu, Z. X.: Relation between ENSO and Precipitation in Shandong . *Yellow River*, **1**, 2007 (in Chinese).

Nakajima, T., Tanaka, M., and Yamauchi, T.: Retrieval of the optical properties of aerosols from the aureole and extinction data, *Appl. Optics*, **22**, 2951–2959, doi: 10.1364/AO.22.002951, 1983.

Nakajima, T. and Tanaka, M.: Algorithms for radiative intensity calculations in moderately thick atmospheres using a truncation approximation, *J. Quant. Spectrosc. Ra.*, **40**, 51–69, doi: 10.1016/0022-4073(88)90031-3, 1988.

Nakajima, T., Tonna, G., Rao, R., Kaufman, Y., and Holben, B.: Use of sky brightness measurements from ground for remote sensing of particulate polydispersions. *Appl. Optics*, **35**, 2672–2686, doi:

10.1364/AO.35.002672, 1996.

Nakajima, T., Yoon, S. C., Ramanathan, V., Shi, G. Y., Takemura, T., Higurashi, A., Takamura, T., Aoki, K., Sohn, B. J., Kim, S. W., Tsuruta, H., Sugimoto, N., Shimizu, A., Tanimoto, H., Sawa, Y., Lin, N. H., Lee, C. T., Goto, D., and Schutgens, N.: Overview of the Atmospheric Brown Cloud East Asian Regional Experiment 2005 and a study of the aerosol direct radiative forcing in east Asia, *J. Geophys. Res.*, 112, D24S91, doi:10.1029/2007JD009009, 2007.

Peel, M. C. and Finlayson, B. L. and McMahon, T. A.: Updated Asian map of the Köppen climate classification system. *Hydrol. Earth Syst. Sci.* 11: 1633-1644, 2007.

Phillips, B. L.: A technique for numerical solution of certain integral equation of first kind, *J. Assoc. Comput. Mach.*, 9, 84–97, 1962.

Pope lli, C.A., Burnett, R.T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., and Thurston, G. D.: Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, *J. Am. Med. Assoc.*, 287(9), 1132–1141, doi: 10.1001/jama.287.9.1132, 2002.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T., Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, *Science*, 294, 2119–2124, 2001.

Rodgers, C. D.: *Inverse Method for Atmospheric Sounding*, World Sci., Singapore, 240, 2000.

Samet, J.M., Zeger, S.L., Dominici, F., Coursac, I., Dockery, D.W., Schwartz, J., and Zanobetti, A.: The national morbidity, mortality, and air pollution study. Part II: morbidity and mortality from air pollution in the United States, Health Effects Institute, Cambridge MA, Research Report 94, 2000.

Sun, K., Liu, H. N., Wang, X. Y., Peng, Z., and Xiong, Z.: The aerosol radiative effect on a severe haze episode in the Yangtze River Delta., *J. Meteor. Res.*, 31(5), 865–873, doi: 10.1007/s13351-017-7007-4, 2017.

Takamura, T., and Nakajima, T.: Overview of SKYNET and its activities, *Opt. Pura Y Apl.*, 37, 3303–3308, 2004.

Tan, S. C., Shi, G. Y., and Wang, H.: Long-range transport of spring dust storms in Inner Mongolia and impact on the China seas. *Atmospheric Environment*, 46, 299-308, 2012.

Tang, J.X., Fu, C. B., Dan, L., Lin X. B.: Analysis on potential sources of air pollutants in Hainan during haze pollution. *Environmental Science and Management*, 06, 2019 (in Chinese).

Tao, J., Zhang, L., Cao, J., and Zhang, R.: A review of current knowledge concerning PM 2.5 chemical composition, aerosol optical properties and their relationships across China. *Atmospheric Chemistry*

- and *Physics*, 17(15), 9485-9518, 2017.
- Twomey, S.: On the numerical solution of Fredholm integral equations of the first kind by the inversion of the linear system produced by quadrature, *J. Assoc. Comput. Mach.*, 10, 97–101, 1963.
- Turchin, V. F. and Nozik, V. Z. : Statistical regularization of the solution of incorrectly posed problems, *Izv. Atmos. Ocean. Phys.*, 5, 14–18, 1969.
- Uchiyama, A., Yamazaki, A., Togawa, H., and Asano, J.: Characteristics of Aeolian dust observed by sky-radiometer in the Intensive Observation Period 1 (IOP1), *J. Meteor. Soc. Japan*, 83A, 291–305, doi: 10.2151/jmsj.83A.291, 2005.
- Wang, S. X., Zhao, B., Cai, S. Y., Klimont, Z., Nielsen, C. P., Morikawa, T., Woo, J. H., Kim, Y., Fu, X., Xu, J. Y., Hao, J. M., and He, K. B.: Emission trends and mitigation options for air pollutants in East Asia. *Atmospheric Chemistry and Physics*, 14(13), 6571-6603, 2014.
- Wang, Z. W., Yang, S. Q., Zeng, Q. L., and Wang, Y. Q.: Retrieval of aerosol optical depth for Chongqing using the HJ-1 satellite data, *J. Meteor. Res.*, 31(3), 586 – 596, doi: 10.1007/s13351-017-6102-x, 2017.
- Wang, Z. Z., Liu, D., Wang, Z., Wang, Y. J., and Khatri, P.: Seasonal characteristics of aerosol optical properties at the SKYNET Hefei site (31.90°N, 117.17°E) from 2007 to 2013, *J. Geophys. Res.*, 119, 6128–6139, doi:10.1002/2014JD021500, 2014.
- Wen, L., Chen, J.M., Yang, L.X., Wang, X.F., Xu, C.H., Sui, X., Yao, L., Zhu, Y.H., Zhang, J.M., Zhu, T., and Wang, W.X.: Enhanced formation of fine particulate nitrate at a rural site on the North China Plain in summer: The important roles of ammonia and ozone, *Atmospheric Environment*, 101, 294-302, 2015.
- Wu, D., Wu, C., Li, F., and Chen, H.: Air pollution episode in southern China due to the long range transport of coarse particle aerosol. *China Environmental Science*, 31(4), 540-545, 2011(in Chinese).
- Yang, Y. R., Liu, X. G., Qu, Y., An, J. L., Jiang, R., Zhang, Y. H., Sun, Y. L., Wu, Z. J., Zhang, F., Xu, W. Q., and Ma, Q. X.: Characteristics and formation mechanism of continuous hazes in China: A case study during the autumn of 2014 in the North China Plain, *Atmos.Chem. Phys.*, 15, 8165-8178, doi: 10.5194/acp-15-8165-2015, 2015.
- Yin, Y., Zhu, D., Tang, W.W., Martini I. P.: The application of GPR to barrier-lagoon sedimentation study in Boao of Hainan Island. *Journal of Geographical Sciences*, 12(3), 313-320, 2002.
- Zhao, B., Wang, P., Ma, J.Z., Zhu, S., Pozzer, A., and Li, W.: A high-resolution emission inventory of

primary pollutants for the Huabei region, China. *Atmos. Chem. Phys.*, 12, 481-501, 2012.

Zhao, B., Liou, K.-N., Gu, Y., Jiang, J. H., Li, Q., Fu, R., Huang, L., Liu, X., Shi, X., Su, H., and He, C.: Impact of aerosols on ice crystal size, *Atmos. Chem. Phys.*, 18, 1065-1078, DOI 10.5194/acp-18-1065-2018, 2018a.

Zhao, B., Zheng, H., Wang, S., Smith, K. R., Lu, X., Aunan, K., Gu, Y., Wang, Y., Ding, D., Xing, J., Fu, X., Yang, X. D., Liou, K. N., and Hao, J.M.: Change in household fuels dominates the decrease in PM_{2.5} exposure and premature mortality in China in 2005–2015. *Proceedings of the National Academy of Sciences*, 115(49), 12401-12406, 2018b.

Zhao, B., Jiang, J. H., Diner, D. J., Su, H., Gu, Y., Liou, K.-N., Jiang, Z., Huang, L., Takano, Y., Fan, X.H., and Omar, A. H.: Intra-annual variations of regional aerosol optical depth, vertical distribution, and particle types from multiple satellite and ground-based observational datasets, *Atmos. Chem. Phys.*, 18, 11247–11260, doi:10.5194/acp-18-11247-2018, 2018c.

Zhu, D., Yin, Y., Martini, I. P.: Geomorphology of the Boao coastal system and potential effects of human activities - Hainan Island, South China. *Journal of Geographical Sciences*, 15(2):187-198, 2005.