Dear Editor, dear reviewers,

Many thanks for the valuable comments, which help to improve the quality of this paper. The detailed replies are addressed below point by point.

As below, I would like to clarify some of the points raised by the reviewers. We hope that the reviewers and the editors will be satisfied with our responses to the 'comments' and the revisions for the original manuscript.

Best regards,

Yahui Che on behalf of all authors

2019-5-15

Response to reviewer 1:

Interactive comment on "Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data" by Yahui Che et al.

Andrew Sayer (Referee) andrew.sayer@nasa.gov

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I am writing this review under my own name (Andrew Sayer) as I have worked with all the satellite instruments the authors use here (although on different algorithms for the ATSRs/AVHRRs), am one of the people who developed the MODIS C6.1 aerosol products used, and am familiar with the authors' work on this topic from their previous studies and discussions at conferences.

This paper seeks to assess whether the gap in aerosol optical depth (AOD) data between the end of the AATSR record in 2012 and start of the SLSTR mission in 2016 (using an algorithm developed by co-author de Leeuw's group) can be bridged with an AVHRR-based algorithm developed by the other authors. Since we are now at the stage where we have multiple satellite records of decent length and quality, it is reasonable to ask how these might be best combined and what sort of consistency can be achieved. So on that front the study is relevant and important. The authors focus on north-eastern China, and in particular two AERONET sites in the Beijing area.

In all honesty the choice of AVHRR and the Beijing region to attempt to bridge the AATSR/SLSTR records seems to be guided in part by the authors' own created data sets and home institution, as the AVHRR sensor is perhaps the least capable of satellite instruments which could have been chosen to fill in this gap, and aerosols in China are complicated so perhaps not the place in the world you'd want to start developing a framework for combining records. The concepts behind the ATSR and AVHRR AOD retrieval algorithms used, and their sampling limitations, are also quite different, which would also affect the expected level of consistency. For example, you might expect a MISR-based algorithm to be a better bridge because MISR and the ATSRs are all multiangle, narrow-swath sensors. I suppose I am left asking: why did the authors try to answer question "how well can I bridge the AOD gap between AATSR and SLSTR in north-eastern China using this AVHRR data set?" rather than the more general questions "how well do available satellite records allow us to create a long-term AOD record over north-eastern China?" or "how should we combine different satellite aerosol records?". This analysis is a step in that direction which can provide input to answering those general questions, so I think it does have value. I can understand some scientific rationale behind this because as the authors point out the AVHRRs are the longest available sensor series (going back to around 1980), and that part of the world has undergone a lot of change in that period with considerable aerosol sources. So scientifically it makes some sense to look at this region and sensor. However I would have liked to see that aspect emphasised more in the paper: the title implies a bigger scope, and the bulk of the analysis in the paper sticks to the post-2000 era.

My overall recommendation is for major revisions; I would like to review the revised version. There is value in this analysis but I think some things need clarification, some need extention, and more

big-picture guidance about the insights the authors have got would be useful for readers planning follow-on studies on this topic. Some specific comments and suggestions relating to these general comments are below:

Re: Thank you for your recommendation and give us a chance to improve the manuscript. We have addressed these points in the revised manuscript.

Title: I suggest adding "over north-eastern China" or similar after "Climate Data Record", to better fit the scope and content of the paper. The current title implies a more large-scale analysis. Re: We revised the title by adding "over North-Eastern China from 1987 to 2012" to indicate both time and regions.

Introduction: SLSTR was launched in 2016 and this paper is largely about bridging the 2012-2016 gap. However, no SLSTR data are shown in the study. Somewhere in here, could the authors add a few statements about the current status of SLSTR AOD data? If there is a SLSTR product in development, it would make sense to wait until that can be included in the analysis (even if it is preliminary). That way we could answer the question of whether AVHRR can bridge the gap, by seeing how consistent the results are with actual SLSTR retrievals. If there is no SLSTR product in development then ok, but this should be stated, and the premise of the paper becomes more questionable (why try to bridge if there is nothing at the other end of the bridge?). On a related note, the AVHRR data processed here end in 2014 (although the AVHRRs are still flying) so would not be able to bridge the gap anyway unless the authors extend the processing period.

Re: Thanks for this remind. To the current, SLSTR data are not available. Hence, we switched to extend ATSR data set back to 1980s, no longer focus on bridging gap between AATSR and SLSTR.

Introduction: As noted in my general comments, I'd like to see more discussion of the bigger picture of why we want to combine satellite records, how it has been done, what the challenges are, and why these sensors and this region were chosen as an example.

Re: Referee 3 also mentioned this point. We switched focus on making an extention back to 1980s for ATSR data set, no longer pay attention to bridge the gap of AATSR and SLSTR. Corresponding revisions have been made in new version of manuscript.

Page 6 line 4: I suggest citing a paper here (there are lots of AVHRR aerosol/cloud/surface papers which give instrument descriptions) rather than an ESA webpage, especially for a non-ESA sensor.

Re: Here we update the citation to "NOAA KLM user's guide" from NOAA official website.

Page 6, lines 15-25: Surface reflectance modelling is one of the largest error sources for AOD retrieval, especially for a sensor like AVHRR which has only a few, and quite broad, spectral bands. The authors' algorithm relates 3.75 micron to 0.64 micron reflectance using an empirical relationship which is a function of surface type. The surface cover type used is obtained from the MODIS MCD12C1 product. This is static for a given location, therefore, if the surface cover changes during the period, the AOD retrieval error will change through time. This is not discussed in the paper but is quite important if the goal is to make a data record going back to the 1980s, as we know there has been a lot of industrial station since then. Basically we know that surface type has changed over parts of this region, so we know that this key algorithm assumption has been violated.

Yet I did not see this discussed in this paper, or in the previous algorithm papers cited here. There should be some discussion and some quantification of the effects of an incorrect surface type classification on the retrieved AOD. (For example, if you use the model for type A instead of type B, how much does the retrieved AOD change, and how systematic is that?) It is also not clear that this surface model can account for the (non-negligible) differences in the spectral response functions between the different sensors, which could result in systematically different reflectances observed for the same underlying surfaces. If these aren't addressed then it undercuts the idea of trying to get a long term data record.

Re: This version of AVHRR AOD data set does have some limitations or disadvantages. As you know, it is very difficult to retrieval AOD when there only one visible band, besides there is no enough prior knowledge in the early time. This paper focus on the analysis of the first version of AVHRR AOD results but not the algorithm itself. Now we are trying to make more detailed validation and improve this algorithm. However, the newly version of long term AVHRR AOD data set is not available now.

Page 6 line 30: NOAA15 says 199-2002, I guess this should be 1998 or 1999 rather than 199? Also, TETOP-A should read METOP-A.

Re: The data from NOAA-15 is from 1992 to 2002, so it should be 1999. These two typos have been revised in new version of manuscript.

Page 8 line 14 and later: The authors sometimes refer to "radiance-derived AOD", sometimes "solar radiation", and sometimes "broadband extinction", when describing one of the ground-based data sets. These phrases, so far as I can tell, all refer to the same data set, but they do not have any words in common. I suggest keeping the terminology consistent to make things clear. My personal preference would be to say "broadband solar radiation" but I'd leave that up to the authors. This refers to some figure captions as well as the body text.

Re: Here we use "BEM (broadband extinction method) AOD" throughout new manuscript, as it does in Dr. Ling Sun's paper. https://doi.org/10.1016/j.atmosenv.2015.08.042

Page 9 line 5: There is now an AERONET version 3 paper published, which could be cited here. https://www.atmos-meas-tech.net/12/169/2019/

Re: Thanks. We have citied this paper in new version of manuscript.

Page 10 line 21: This says that the AVHRR retrievals are available from 1983-2014. But from page 6, only data back to 1987 are used in this study. Why not include the first four years too, since the authors state they processed the data?

Re: All instruments we selected in this paper are onboard satellites along with descending orbit. For years from 1983 to 1986, AVHRR raw data are not consistent, data only from 1986.11 to 1986.12 (from NOAA-10), 1985.07 to 1985.10 (from NOAA-8), 1984.05 to 1984.06 (from NOAA-8) and 1984.07 (from NOAA-6), and 1983.01 to 1983.03 (from NOAA-6) are available. So we abandon data from 1983 to 1986. This is stated in new version of manuscript.

Figure 2: We know that AOD changes a lot in different seasons. We also know that satellite biases are affected by geometry, AOD/aerosol type, and surface reflectance, which also change a lot in

different seasons. We also know that sampling is related to cloud and snow cover, which also change a lot in different seasons. We know that all these factors can affect different algorithms in different ways. Given that, I do not find multiannual means as presented in this Figure to be useful, as we have no idea which factors are contributing to the differences observed here. I suggest changing this to show multiannual seasonal composites instead of overall multiannual means. I feel that will be more useful and allow for more insight about these confounding factors. Otherwise the authors need to provide a justification why presenting the data in this way is useful, given these issues.

Re: Thanks, seasonal characteristics of differences and spatial distribution are really helpful for understanding AOD retrieval algorithms and data sets. Seasonal AOD and AOD differences have been added into new version of manuscript.

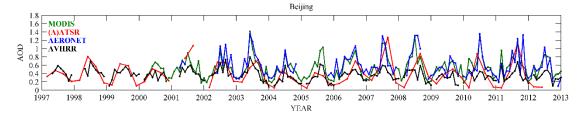
Page 12, line 23: should be changed to "to MODIS Dark Target over land", for completeness. This expression does not apply to the Deep Blue or merged product (which is what is actually used here). Re: This has been revised in new version of manuscript.

Page 13, lines 28-30: If my reading is correct, the authors are saying that the MODIS C6.1 validation results obtained here have different error characteristics than the MODIS C6.1 validation results over the same region obtained by Sogacheva et al (2018). Is that correct? If so, it seems surprising, so needs to be examined in more detail. Is there an error in one of the studies? Where does the difference come from? If it is a result of a different sampling period or sites, that is important to discuss, as it indicates that the regional validation is not robust and therefore that we cannot take these results as representative. This should be clarified in the text.

Re: Yes, that is correct, but the region is not e exactly the same. Moreover, the difference is that we used data from both AERONET and CARSNET in validation while Sogacheva et al (2018) only used AERONET data in validation. This is why there are different error characteristics. As shown by fig.1 in manuscript, all AERONET sites are in or around Beijing where main surface types are urban, croplands or mixed forest and aerosol sources are mainly anthropogenic, on the contrary CARSNET sites distribute in more regions in study area especially in sparsely vegetated regions where the surface and aerosol circumstances are significantly different. Hence, the conclusions are with small differences. This has been clarified in the new version of manuscript.

Figures 4-6 (and 9, 10): I find these a little hard to interpret. We are trying to see how consistent the data sets are with AERONET and each other, yet by plotting all of them on separate panels it makes harder to see how consistent they are. I suggest plotting all the data sets on a single panel instead, so we can directly compare them. I realize that the ATSR data shown are seasonal while the others are monthly, due to sampling limitations. In that case one option is just to plot everything (including AERONET) as a seasonal rather than monthly time series. This would also remove the issue of different expected extreme which the authors mention in captions.

Re: Seasonal comparison rather than monthly comparison in time series is definitely more concise for reading, but that will lose some details. Hence, we just keep monthly time series for each except ATSR. On this basis, if we put everything into one plot, this plot will be too messy to read like following figure.



The focus of this paper is to investigate consistency of ATSR and AVHRR, while AERONET and MODIS are chosen as reference data. Hence, we make comparison of MODIS and AERONET separately to see the performance of MODIS. When it is confirmed, time series comparison will be made with AERONET or MODIS as reference separately. Please refer to fig. 4, 5, 9, and 10 in new version of manuscript. Those time series are looking much better than before.

Figures 7, 8: these are interesting, but I feel they should be expanded to make the paper more complete. They show that the monthly solar broadband-based AOD is a reasonable proxy for AOD from AERONET or MODIS. But why not also include scatter plots like figure 7 for AVHRR and AATSR? And why not include the full time series, as well as ATSR data, in figure 8? Basically the paper is about a 1983/7 and onward AOD data set over Beijing, but nowhere is a full time series of these data actually shown in the paper.

Re: As last comments, we didn't put everything into one full time series plot. Instead we made two time series, one is to validate BEM AOD and the other is to be as reference to validate AVHRR and ATSR. Please refer to fig. 6 and 8 in new version of manuscript.

Conclusions: The paper also builds on other recent work by the de Leeuw group which assessed a combination of the ATSR and MODIS sensors to get a longer-term AOD over China. This is mentioned a few times early on, but I think the conclusion should be expanded to put these studies in perspective with each other, and give some more specific thoughts/suggestions on how best to combine data records. Overall this paper has some good points but suffers from not always following through to give the bigger picture (i.e. when you have finished your investigation of the long-term climate data record, as promised in the title, what does it actually look like)?

Re: Together with other two reviewers' comments, we added more conclusions.

Response to reviewer 2:

Interactive comment on "Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data" by Yahui Che et al.

Anonymous Referee #2

Received and published: 28 February 2019

Review of "Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data" for Atmospheric Measurement Techniques. This paper try to discuss the feasibility of using AVHRR to continue the aerosol optical depth (AOD) record from AATSR ending in 2012 to SLSTR starting at 2016 over Beijing-Tianjing-Hebei region. The study is relevant and the potential product will benefit the aerosol community. However, there are some major issues that need to be addressed before it is suitable for publishing.

Re: Thank you for affirmation and suggestions. We have addressed these points in the revised manuscript.

1. The reason author chooses to use AVHRR is because not only can this data bridge the gap between AATSR and SLSTR but also it can extend the data record to 1983. This idea is presented in introduction, but there is only one plot Figure 8 shows the AVHRR data before 2000. All other analyses are focused 2000 to 2012. I think if we only consider this 2000-2012 period of time, there are a lot more aerosol products that can be used with much lower uncertainties. Thus, more analyses are needed to understand AVHRR through the entire data record or empirically correct AVHRR data to make it more suitable for a long term data record.

Re: As you suggest, AHVRR is not a good choice as there are some aerosol products with lower uncertainties like MODIS or MISR aerosol product. However, AVHRR is almost the only choice to look back to 1980s with expected accuracy. Correspondingly, we change the focus on making an extention (A)ATSR aerosol data record back to 1980s using AVHRR data. We will not talk about bridge of AATSR and SLTSR in this paper. Please refer to the introduction in new version of manuscript.

2. This is a paper about continue data from AATSR to SLSTR. But I didn't see a session in data talking about SLSTR.

Re: We change the primary purpose of this paper to extention (A)ATSR aerosol data record back to 1980s. Hence, we will not talk about SLSTR.

3. The author uses a very large portion of paper introducing aerosols and their facts. It is really not to the point of this article. Please make the introduction more concise.

Re: This is also mentioned by Dr. Andrew Sayer (referee 1). Please refer to the new version of manuscript in which the introduction is re-written.

4. To me it makes more sense to validate the radiance retrieved AOD against AERONET. Or maybe against MODIS to show the sampling bias. Then rely on radiance retrieved AOD to validate everything else consistently from 1983 to current.

Re: The validation of this "BEM (broadband extinction method) AOD" (the radiance retrieved AOD)

has already done co-author Dr. Ling Sun of by in her paper https://doi.org/10.1016/j.atmosenv.2015.08.042. The primary purpose of this paper has been switched to make an extention from 1987 to 1997 from ATSR. Hence, we adjust radiance AOD related time series from 1987 to 2012 as whole-time range (please refer to fig.6 and 8 in new version of manuscript). Here, we only validate AVHRR AOD with radiance retrieved AOD as the data volume for monthly ATSR AOD is too small which could lead to severe bias.

5. The title doesn't indicate the study region.

Re: The title has been revised as "Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data over North-Eastern China from 1987 to 2012".

Response to reviewer 3:

Interactive comment on "Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data" by Yahui Che et al.

Anonymous Referee #3

Received and published: 16 March 2019

This manuscript compares aerosol retrievals from 3 different sensors (MODIS, ATSR, and AVHRR) with ground based validation data (including AERONET) over a 10x10 degree box centered on eastern China. The goal is a worthy one, the creation of a continuous climate data record of satellite-retrieved aerosol optical depth from the early 1980's until the present day period. I believe this work can be published if the authors are willing to substantially change the manuscript. Most of my comments are embedded within the PDF attached, but I will summarize a few points.

Re: Thank you for affirmation and the constructive comments. We have addressed these points in the new version of manuscript.

- 1) As was mentioned by the other two reviewers, where is the SLSTR data? If this data is not available, this manuscript can still add value, but not much (in its current form).
- Re: The focus of this paper has been switched to extent ATSR data set back to 1980s. The content about SLSTR data is deleted in the new version of manuscript.
- 2) MISR shares a lot of similarities to ATSR including: swath size, multi-angle viewing, equatorial crossing time, and algorithm heritage (over-land). Additionally, I expect that the error statistics for MISR (over this region) are quite a bit better than for any other sensor used in this paper. In fact, given MISR's very long data record (2000–>2019 and counting), its similarities with ATSR, its overlap with *both* ATSRs *and* SLSTR, I think it makes much more sense to use MISR to stitch together the ATSRs and SLSTR. Once those two datasets are harmonized with MISR (globally, not for one region), I would then look back and compare with AVHRR (globally, or at least using all regions available).

Re: Thanks for this suggestion. MISR data is possibly a better choice to bridge the gap of AATSR and SLSTR data. As mentioned above, the content SLSTR data are deleted, and we have to switch the focus of this paper on period from 1987 to 2012.

3) Please find a way to make this work much more global. A 10x10 degree region is not a very useful climate data record, especially in a region with so much dust and pollution transport. Additionally, the authors could show consistencies and discrepancies with other sensors via a map of gridded correlations and differences (using seasonal AODs, compared with other sensors).

Re: Thank you for your suggestions. We start from the small regions as we only have this region AVHRR long-term AOD till now. Besides, this region with complex aerosol types helps us to analyze the effectiveness of AOD products in complex situations, because it's easy for everyone to be very accurate in simple situations. Correlation maps are not the best choice as the data volume of seasonal maps are with big differences. Hence, we plot seasonal difference maps for three products.

4) As a third party (I work with MISR data) with no stake in any of these instruments (at least data from the ones presented), it seems pretty clear from this small dataset that MODIS provides the best available AOD here (by far). One (or more) of three things is going on here: (1) AATSR's aerosol retrieval algorithm is inferior to MODIS, (2) AATSR's sample size in this region is so small as to border on the irrelevant, or (3) the region selected is so small that regional biases in the algorithm dominate your observed errors. If (1), I have to wonder why bother stitching together AOD from ATSR and AATSR with SLSTR (and AVHRR) at all? Even though ATSR, AATSR, and SLSTR all lack a blue band (which will significantly degrade performance over brighter regions), this should be compensated by the additional view angle. If the current algorithm is insufficient, maybe a new one should be developed. Otherwise, if MODIS truly gives better performance, just create the CDR using MODIS, AVHRR, and VIIRS, which would be easier anyways. If (2) or (3) see point 3) above, you need more data.

Re: The focus of this paper has been switched to make an extention of ATSR back to 1980s, and the study area is still limited to north China before newly AVHRR data are produced. The performance of ATSR is inferior to MODIS over small region but they are very close when study area is extended to whole China region as described in Sogacheva et al (2018). Besides, ATSR could provide data set before 2000. Our following work is to produce AVHRR data set over whole China.

Reply to comments in supplement:

- 1. Page 1, line 6, AOD>0.6 is not uncommon for the Beijing region in winter either. Re: We revised this in the new version of manuscript.
- 2. Page 3, line 11, delete "and thus restore nature"Re: We rewrote introduction in new version of manuscript in which this line was deleted.
- 3. Page4, line 22, Why choose only China if Europe is also available with this dataset? I would expect that Europe may have had some validation data going further back than the validation data available over China.

Re: Our following work is to produce AVHRR AOD data set over whole China region and we have BEM AOD (boardband extinction method) from at least ten sites. Hence, in this paper we focus on north China, but we will consider Europe in next paper.

4. Figure 1, This figure needs to have land cover included on the colormap. We should not have to scroll up and down.

Re: Color codes have been replaced by the name of land covers.

- 5. Page 5, line 22, Any reason MISR is not included here? Of the sensors listed here, it probably compares most favorably with validation in the region. The latest version (23) of the product also has an improved spatial resolution (17.6-->4.4 km), and performs noticeably better at high AOD. Re: MISR is really a good instrument for producing aerosol data set. The current purpose of this paper is to extent AOD back to 1987. When SLSTR data are available, MISR is possibly the best choice to bridge the gap of AVHRR and SLSTR. But this will be considered in another study.
- 6. Page 6, line 19, Sea spray extinction probably never exceeds 0.01 at 3.75 microns (and is probably

much smaller).

Re: We only consider the land surface, and sea is not included in this study of AVHRR AOD.

7. Page 7, line 6, I am curious, is this 55 degrees from the surface normal, or is boresight angle 55 degrees?

Re: 55 degrees are from nadir to forward. We have added an explanation of this in the new version of manuscript.

8. Page 7, line 9, Similar swath to MISR (400 km), similar crossing time to MISR as well.

Re: Thanks for this remind and we will use MISR data to bridge the gap between AATSR and SLSTR when SLSTR data is ready.

9. Page 7, line 11, MISR's shape-similarity algorithm (part of the land algorithm) is probably based on this heritage.

Re: Thanks again.

10. Page 11, line 6, It would be relatively simple to perform a correlation between the different sensors. Additionally, a correlation (and RMSD) map based on seasonal mean AODs could be generated as well. This could give a very good indication on spatial agreement.

Re: We have plotted seasonal AOD difference maps instead of correlation maps as the data volume in averaging seasonal AOD for ATSR is much less than the other two.

11. Page 11, line 12, delete "overall somewhat"

Re: It has been deleted.

12. Page 12, line 11, For MODIS or ATSR?

Re: It is the difference between MODIS and ATSR.

13. Page 13, line 12, Hence the importance of using data from all available regions (including Europe). This will also greatly increase the amount of data available from AATSR.

Re: The presented paper focuses on data sets over north China, but our following research will be extended to whole China regions so that the data volume of ATSR will not be a problem. We will consider Europe as well.

14. Page 13, line 13-15, replaced by "an adequate retrieval over bright surfaces".

Re: This line has been replaced by "an adequate retrieval over bright surfaces".

15. Figure 3, as a third party with no stake in any of these instruments, it seems pretty clear (from this small dataset) that MODIS provides the best available AOD here (by far). One (or more) of three things is going on here: AATSR's aerosol retrieval algorithm is inferior to MODIS, AATSR's sample size in this region is so small as to border on the irrelevant, or the region selected is so small that regional biases in the algorithm dominate your observed errors.

Re: The same as (4).

16. About qualities of figures.

Re: We will upload figures with high quality with a resolution of at 300dpi separately.

Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data <u>over</u>

North-Eastern China from 1987 to 2012

Yahui Che^{1,7}, Jie Guang¹, Gerrit de Leeuw^{2,43}, Yong Xue^{1,34}, Ling Sun⁵, and Huizheng Che⁶

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Abstract. Satellites provide information on the temporal and spatial distributions of aerosols on regional and global scales. With the same method applied to a single sensor all over the world, a consistent data set is to be expected. However, the application of different retrieval algorithms to the same sensor, and the use of a series of different sensors may lead to substantial differences and no single sensor or algorithm is better than any others everywhere and at any time. For the production of long-term climate data records, the use of multiple sensors cannot be avoided. The Along Track Scanning Radiometer (ATSR-2) and the advanced ATSR (AATSR) Aerosol Optical Depth (AOD) data sets have been used to provide a global AOD data record over land and ocean of 17-years (1995-2012), which is planned to be extended with AOD retrieved from a similar sensor, i.e. the Sea and Land Surface Temperature Radiometer (SLSTR) which flies on Sentinel 3A launched in early 2016. However, this leaves a gap of about 4 years between the end of the AATSR and the start of the SLSTR data records. To fill this gap, and to To investigate the possibility to extend the ATSR data record to earlier years, the use of an AOD data set from the Advanced Very High Resolution Radiometer (AVHRR) is investigated. AOD data sets used in this study were retrieved from the ATSR sensors using the ATSR Dual View algorithm ADV v2.31 developed by Finnish Meteorological Institute (FMI), and from the AVHRR sensors using the ADL algorithm developed by RADI/CARCAS. Together these data sets cover a multi-decadal period (19873-2014). The study area includes two contrasting areas, both as regards aerosol content and composition and surface properties, i.e. a region over North-East (NE) China encompassing a highly populated urban/industrialized area (Beijing-Tianjin-Hebei) and a sparsely populated mountainous area.

Ground-based AOD observations available from ground-based sunphotometers un photometer AOD data in AERONET and CARSNET are used as reference, together with broadband extinction methods (BEM) radiation derived AOD data at Beijing to cover the time before sun photometer observations became available in the early 2000's. In addition, MODIS-Terra C6.1

AOD data are used as reference data set over the wide area where no ground-based data are available. All satellite data over the study area were validated versus the reference data, showing the qualification of MODIS for comparison with ATSR and AVHRR. The comparison with MODIS shows that AVHRR performs better that ATSR in the north of the study area (40 °N), whereas further south ATSR provides better results. The validation versus sun photometer AOD shows that both AVHRR and ATSR underestimate the AOD, with ATSR failing to provide reliable results in the winter time. This is likely due to the highly reflecting surface in the dry season, when AVHRR-retrieved AOD traces both MODIS and reference AOD data well. However, AVHRR does not provide AOD larger than about 0.6 and hence is not reliable in the summer season when high AOD values have been observed over the last decade. In these cases, ATSR performs much better, for AOD up to about 1.3. AVHRR-retrieved AOD compares favourably with radiance-derived AOD, except for AOD higher than about 0.6. These comparisons lead to the conclusion that AVHRR and ATSR AOD data records each have their strengths and weaknesses which need to be accounted for when combining them in a single multi-decadal climate data record.

1 Introduction

Aerosol particles are important atmospheric constituents, which play significant roles in many processes such as atmospheric chemistry, the absorption and scattering of solar radiation, and the lifetime of cloud and precipitation systems (Boucher et al, 2013; Koren et al., 2014; Guo et al., 2014; 2016a; 2018). Aerosol particles have an adverse effect on human health and are responsible for 7 million premature deaths annually over the whole world (WHO, 2018). Processes involving aerosols and their effects depend on the chemical and physical properties of the aerosol particles which in turn are determined by sources of directly emitted particles, the formation of aerosols from precursor gases (and thus the sources of these gases), the transformation of these particles during chemical and physical processes in the atmosphere and their removal by wet or dry deposition (cf. Seinfeld and Pandis, 1998, for a comprehensive treatment of aerosol processes). Observations of the concentrations of trace gases and aerosols in China are publicly available since several observational networks have been established, such as CARE China (Xin et al., 2015), and the NASA's AERONET (AErosol RObotic NETwork) (Holben et al. 1998) (with observations mainly in the east of China), CARE-China (Xin et al., 2015), the Chinese CARSNET (Chinese Aerosol Remote Sensing Network (CARSNET)) (Che et al., 2009; 2015) and SONET (Sun-sky radiometer Observation NETwork) (Li et al., 2018), all of which provide data across the whole country. However, most of these observations started in the last decade and very few, if any, historical data on large scale are available for the construction of the long time series needed to show the evolution of pollutant concentrations over many years and analyse the effects of different contributions. Here, satellite data may offer a solution. The most common satellites used for the observation of trace gases and aerosols offer information since the beginning of the 21st century and, by combining the information from different instruments, time series encompassing two decades can be constructed (de Leeuw et al., 2018, Sogacheva et al., 2018b). Satellite information has been used together with model simulation to analyse the effects of natural and anthropogenic contributions on the concentrations of trace gases and aerosols (Kang et al., 2018). In another study combining satellite data with ground-based observations, the role

of precursor gases, in particular VOCs, and photochemical reactions in the formation of aerosols $(PM_{2.5})$ was revealed (Bai et al., 2018).

In the second half of the 20th century the adverse effects of the precursor gases like SO₂ and NO₂, and the induced aerosols on climate, air quality, and health were recognized and reduced by effective measures in developed countries. These led to the reduction of air pollution in developed countries, in particular in North America and Europe (Guerreiro et al., 2014), but in developing countries with increasing industrial activity and urbanization the concentrations continued to increase (Hao et al., 2000). As an Ecxample, isn China where the concentrations of pollutants have increased over the years and are amongst the highest in the world. Recent publications show the effect of policy measures on the reduction of the most polluting trace gases

10 SO₂ and NO₂ (van der A et al., 2017), which, as precursor gases, also affect the concentrations of aerosols. In particular, the emissions of SO₂ were reduced as part of the 11th Five-Year Plan (2006-2010) (Zheng et al., 2018), but the emissions of NO₂ continued to increase (e.g., van der A et al., 2017) until the 12th Five-Year Plan (2011-2015). Large emission reductions were achieved after 2013 when the Clean Air Action was enacted and implemented and the NO₂ concentrations decreased (Zheng et al., 2018). Starting from 2011, aerosol concentrations decreased in China as shown, e.g., from satellite observations of the aerosol optical depth (AOD) (Zhang et al., 2017, Zhao et al., 2017, de Leeuw et al., 2018, Sogacheva et al., 2018b). However, early policies before 2005 on reducing pollution emissions are not effective, as shown from sparse ground-based observations (Jin et al., 2016). Long term satellite retrievals may offer a better-solution on a (larger spatial scale) on to explaining observe how air pollutions control policies worked and presenting air pollution changeds.

The Along Track Scanning Radiometer (ATSR-2), a dual view instrument, was launched in 1995 on the European Space Agency (ESA) satellite ERS-2 and provided data until 2003, and was one of the earliest satellite instruments used to retrieve Aerosol Optical Depth (AOD) quantitatively (Flowerdew and haigh, 1996; Veefkind et al., 1998). Its successor, the Advanced ATSR (AATSR) is a similar instrument launched in 2002 on the ESA platform ENVISAT, which was lost in April 2012. The AOD over China from ATSR-2 and AATSR is consistent (Sogacheva et al., 2018a) and hence, together these instruments provide a 17-year AOD time series, 1995-2012 (Popp et al., 2016, de Leeuw et al., 2018). Combining ATSR and MODIS, 22-year AOD measurements were constructed, showing how the AOD increased until about 2006, and then clearly decreased since 2011 over China (de Leeuw et al., 2018, Sogacheva et al., 2018b). MODIS is approaching the end of its life time, but the AOD time series will be extended with NPP/VIIRS (cf. Levy et al., 2015). The AATSR time series will be extended with data from the operational Sea and Land Surface Temperature Radiometer (SLSTR) which was launched on Sentinel 3 in February 2016. This leaves a gap of about 4 years between AATSR and SLSTR. This gap could be filled with MODIS data, as described in Sogacheva et al. (2018b) who combined ATSR and MODIS/Terra (C6.1) data to construct an AOD time series 1995-2017However, this time series should be extended especially before the 1990s when great changes have taken place in aerosols in China with the increase of industrialization to meet requirements of aerosol and other communities, this time series needsthis

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is still expected to be extended especially before the 1990s to meet requirements of other communities. The Advanced Very High Resolution Radiometer (AVHRR) onboard the NOAA series satellites series is could be a good choice to longerextend thise ATSR time series as it started observations continuously from 1978 to the present. The-Xue et al. (2017) developed an AOD data set encompassinges only two relatively small areas over Europe and China from 1983 to 2014, but covers the 5 complete period, as opposed to the AVHRR global over land AOD data set recently presented by Hsu et al. (2017) and Sayer et al. (2017) which encompasses several distinct time periods. Hence in this study we focus here on the Xue et al (2017) AVHRR AOD data set over China and compare that with ATSR-derived AOD data to determine its suitability for merging (as done for ATSR/MODIS by Sogacheva et al. 2018b), and thus extending the ATSR data set both-before the ATSR-2 era (and possibly after the AATSR was lost in 2012, although other data sets may be more suitable for to extend to later years as shown in Sogacheva et al. (2018b) for MODIS. So the focus The objective of the current study is to investigate whether this the ATSR AOD data set can also be done extended to earlier years by using the Xue et al. (2017) AVHRR AOD data over China. For comparison of the ATSR and AVHRR AOD data sets, we use , using the AOD data set over land which was recently presented by Xue et al. (2017). The most common instrument used for aerosol retrieval is the Moderate Resolution Imaging Spectroradiometer (MODIS) which was launched on the Terra satellite, in a morning orbit with equator crossing time 15 (descending) at 10:30 local time (LT), in December 1999 and on the Aqua satellite (ascending, equator crossing time 13:30 LT) in May 2002. MODIS thus provides an AOD time series since 2000 and is still operational. In this study we use both ground-based reference data, from AERONET (Holben et al., 1988) and from CARSNET (Che et al, 2009, 2015), and MODIS C6.1 AOD data for comparison and evaluation. These -Reference data and MODIS/Terra data for inter-comparison are not available for the period before 2000 and therefore we also use AOD data derived from broadband radiation measurements using the broadband extinction methods (BEM) derived AOD data (Xu et al., 2015, Guo et al, 2016b) as described in Sect. 2.3.3. Data sets and methods used are presented in section 2. An overview of the data and an evaluation of their quality are presented in section 3, including a comparison of the various data sets. The results are discussed and conclusions are presented in section 4. era until SLSTR

Aerosol particles can be emitted by natural processes such as the interaction between wind and waves which produces sea

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In addition to this large variety of sources, the aerosol concentrations vary strongly with meteorological conditions, which in turn are affected by large scale synoptic conditions and weather systems (Li et al., 2017a; Miao et al., 2017; Yim et al., 2019). The aerosol concentrations also vary with economic development such as industrialization, urbanization and the ensuing policy measures to reduce adverse effects on health and climate. Since the industrial revolution in the 18th century, the emission of air pollutants has been increasing until adverse effects were recognized and measures were developed and implemented to reduce emissions and concentrations of pollutants (e.g., Brimblecombe, 2006). In particular, in the second half of the 20th century the effects of SO2 on forests and lakes, also known as acid rain, was recognized and measures were taken to reduce the SO₂ emissions and thus restore nature. Later, the adverse effects of NO₂ emissions and effects of aerosols, especially fine

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particulate matter, on air quality, health and climate were recognized and reduced. These led to the reduction of air pollution in developed countries, in particular in North America and Europe (Guerreiro et al., 2014), but in developing countries with increasing industrial activity and urbanization the concentrations continued to increase (Hao et al., 2000). Examples are China and India where the concentrations of pollutants are amongst the highest in the world. Taking China as an example, recent publications show the effect of policy measures on the reduction of the most polluting trace gases SO2 and NO2 (van der A et al., 2017), which, as precursor gases, also affect the concentrations of aerosols. In particular, the emissions of SO₂-were reduced as part of the 11th Five Year Plan (2006-2010) (Zheng et al., 2018), but the emissions of NO2 continued to increase (e.g., van der A et al., 2017) until the 12th Five-Year Plan (2011-2015). Large emission reductions were achieved after 2013 when the Clean Air Action was enacted and implemented and the NO2 concentrations decreased (Zheng et al., 2018). Starting from 2011, aerosol concentrations decreased in China as shown, e.g., from satellite observations of the aerosol optical depth (AOD) (Zhang et al., 2017, Zhao et al., 2017, de Leeuw et al, 2018, Sogacheva et al., 2018b). Observations of the concentrations of trace gases and acrosols in China are publicly available since several observational networks have been established, such as CARE-China (Xin et al., 2015), and the NASA's AERONET (AErosol RObotic NETwork) (Holben et al. 1998) with observations mainly in the east of China, the Chinese CARSNET (Chinese Aerosol 15 Remote Sensing Network) (Che et al., 2009; 2015) and SONET (Sun-sky radiometer Observation NETwork) (Li et al., 2018), all of which provide data across the whole country. However, most of these observations were established in the last decade and very few, if any, historical data on large scale are available for the construction of the long time series needed to show the evolution of pollutant concentrations over many years and analyse the effects of different contributions. Here, satellite data may offer a solution. The most common satellites used for the observation of trace gases and aerosols offer information since 20 the beginning of the 21st century and, by combining the information from different instruments, time series encompassing two decades can be constructed (de Leeuw et al., 2018, Sogacheva et al., 2018b). Satellite information has been used together with model simulation to analyse the effects of natural and anthropogenic contributions on the concentrations of trace gases and aerosols (Kang et al., 2018). In another study combining satellite data with ground-based observations, the role of precursor gases, in particular VOCs, and photochemical reactions in the formation of aerosols (PM_{2.5}) was revealed (Bai et al., 2018). 25 In this study we focus on aerosols, and in particular on the AOD observed by satellites. The most common instrument used for aerosol retrieval is the Moderate Resolution Imaging Spectroradiometer (MODIS) which was launched on the Terra satellite, in a morning orbit with equator crossing time (descending) at 10:30 local time (LT), in December 1999 and on the Aqua satellite (ascending, equator crossing time 13:30 LT) in May 2002. MODIS thus provides an AOD time series since 2000 and is still operational. The Along Track Scanning Radiometer ATSR 2, a dual view instrument, was launched in 1995 on the European Space Agency (ESA) satellite ERS 2 and provided data until 2003. The Advanced ATSR (AATSR) is a similar instrument launched in 2002 on the ESA platform ENVISAT, which was lost in April 2012. The AOD over China from ATSR-2 and AATSR is consistent (Sogacheva et al., 2018a) and hence, together these instruments provide a 17-year AOD time series, 1995-2012 (Popp et al., 2016, de Leeuw et al., 2018). Combining ATSR and MODIS, 22-year AOD measurements were constructed, showing the AOD increased until about 2006, and then clear decreased since 2011 over China (de Leeuw et al.,

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2018, Sogacheva et al., 2018b), MODIS is approaching the end of its life time, but the AOD time series will be extended with NPP/VIIRS (cf. Levy et al., 2015). The AATSR time series will be extended with data from the operational Sea and Land Surface Temperature Radiometer (SLSTR) which was launched on Sentinel-3 in February 2016. This leaves a gap of about 4 years between AATSR and SLSTR. This gap could be filled with MODIS data, as described in Sogacheva et al. (2018b) who combined ATSR and MODIS/Terra (C6.1) data to construct an AOD time series 1995-2017. The objective of the current study is to investigate whether this can also be done by using AVHRR data, using the AOD data set over land which was recently presented by Xue et al. (2017). The advantage of using AVHRR data is the long time series which would allow extension back to the earliest AVHRR AOD retrievals available, in this case 1983, and thus construct a time series 1983present. The Xue et al. (2017) data set encompasses only two relatively small areas over Europe and China, but covers the complete period, as opposed to the AVHRR global over land AOD data set recently presented by Hsu et al. (2017) and Sayer et al. (2017) which encompasses several distinct time periods. Hence we focus here on the Xue et al (2017) AVHRR AOD data set over China and compare that with ATSR derived AOD data to determine its suitability for merging (as done for ATSR/MODIS by Sogacheva et al. 2018b), and thus extending both before the ATSR-2 era and after the AATSR era until SLSTR. In this study we use both ground-based reference data, from AERONET (Holben et al., 1988) and from CARSNET (Che et al, 2009, 2015), and MODIS C6.1 AOD data for comparison and evaluation. Reference data and MODIS/Terra data for inter-comparison are not available for the period before 2000 and therefore we also use radiance derived AOD data (Xu et al., 2015, Guo et al, 2016b). Data sets and methods used are presented in section 2. An overview of the data and an evaluation of their quality are presented in section 3, including a comparison of the various data sets. The results are discussed and conclusions are presented in section 4.

20 2 Method

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2.1 Study area

The study area is located over north-eEastern China, i.e. between 110 °and 120 °E and 35 °and 45 °N (Fig. 1), which is divided into two sub-regions by the Taihang Mountain range with the North China Plain (NCP) and large urban agglomerations like Beijing and Tianjin and the Hebei province (together BTH which is among the most populated and fast developing regions in China) to the SE and the mountainous terrain to the NW extending over the Loess Plateau in Shanxi Province and the Inner Mongolia plateau. The Taihang mountain range forms a natural barrier for the transport of air pollution resulting in the frequent accumulation of pollutants and the occurrence of haze over the BTH area and the NCP (e.g., Sundström et al., 2012; Wang et al., 2013). The satellite-derived AOD maps in Fig. 2 show that this line also roughly divides high AOD in the SW of the study area and low AOD in the NW. The background in Fig. 1 is a land cover map showing that the major land cover types in the study area are croplands in the SE and grassland to the NW, which are intersected by mixed forest and closed shrublands as shown in the inset in Fig. 1.

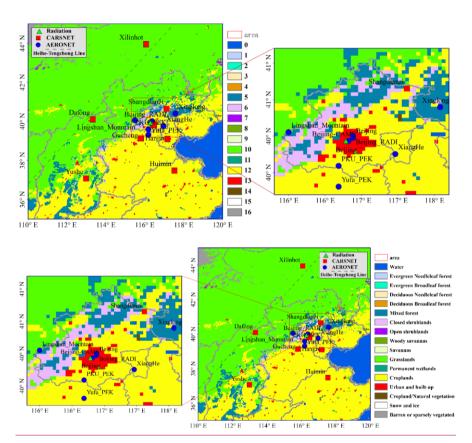


Figure 1: Study area, with the locations of the ground-based reference sites discussed in Sect. 2.3 (CARSNET: red squares, AERONET: blue circles, solar radiation station: green triangle) overlaid on the IGBP land cover map. Land cover is colour coded, see the legend to the right of the map: Water (0), Evergreen Needleleaf forest (1), Evergreen Broadleaf forest (2), Deciduous Roadleaf forest (4), Mixed forest (5), Closed shrublands (6), Open shrublands (7), Woody savannas (8), Savannas (9), Grasslands (10), Permanent wetlands (11), Croplands (12), Urban and built-up (13), Cropland/Natural vegetation mosaic (14), Snow and ice (15) and Barren or sparsely vegetated (16).

2.2 Satellite data

Satellite-retrieved data sets used in this study are satellite-derived AOD data from six radiometers, i.e. AVHRR-1, AVHRR-2, AVHRR-3, ATSR-2, AATSR and MODIS. These data sets are briefly discussed below.

2.2.1 AVHRR

The AVHRR instruments flew on a series of satellites, most of them with an afternoon equator crossing time at 13:40 LT (ascending) (see Xue et al., 2017, for an overview). AVHRR has a swath width of 23992900 km at 833km altitude (Robel and Graumann, 2014https://earth.esa.int/web/guest/missions/3rd_party_missions/current_missions/noaa_ayhrr) and thus provides daily global coverage. The AVHRR sensor was designed for measuring cloud cover and surface temperature, but the observations are also used for the retrieval of AOD over ocean (e.g. Zhao et al., 2008), with only few efforts to retrieve the AOD over land. Recently two AOD data sets retrieved from AVHRR observations over land were published, the first one by Xue et al. (2017) and short after another one by Sayer et al. (2017) and Hsu et al (2017). Xue et al. (2017) used the Algorithm for the retrieval of the aerosol optical Depth over Land (ADL) which was used to produce the continuous data set over China used in the current study. The use of AVHRR data for aerosol retrieval requires a re-calibration of the radiances measured at the top of the atmosphere (TOA) because the sensors have no in-flight calibration. Xue et al. (2017) describe how this was achieved. Major problems in the retrieval of AOD from satellite observations is the effective decoupling of atmospheric and surface effects on the reflectance measured at the top of the atmosphere (TOA), cloud detection and the description of the aerosol properties. In the ADL algorithm a cloud mask is applied based on the Clouds from AVHRR (CLAVR, Stowe et al., 1991, 1999) and in the retrieval six aerosol types are used as proposed by Govaerts (2010). For cloud-free pixels, and after application of gas absorption corrections as described in Xue et al. (2017), the land surface reflectance in the AVHRR channel 1 (0.64 µm) is parameterized in terms of the measured reflectance in Channel 3 (3.75 µm), with coefficients which are functions of the NDVI and scattering angle. The aerosol extinction at 3.75 µm is assumed to be negligible (which may not be true in the presence of coarse aerosol particles such as for desert dust-or sea spray aerosol). It is noted that the TOA reflectance at 3.75µm is composed of solar radiation reflected by the land/atmosphere system and Earth radiation. The reflected part is estimated by using the method proposed by Allen et al. (1990). For each land cover type, as determined following the International Geosphere-Biosphere Programme (IGBP) land cover classification, a different parameterization of the 0.64 µm reflectance was developed. The IGBP land cover information used is the MODIS MCD12C1 product obtained from https://lpdaac.usgs.gov/dataset_discovery/modis/modis_products_table/mcd12c1, and is described in the MOD12 product ATBD (https://modis.gsfc.nasa.gov/data/atbd/atbd_mod12.pdf).

In the implementation of the ADL algorithm, the land surface reflectance is coupled to a radiative transfer model which includes individual parameterizations of the direct, single-scattered and multiple-scattered radiances. Thus a model is developed for the TOA reflectance which is solved by optimal estimation. For more detail, see Xue et al., (2017). AOD is retrieved at a wavelength of 0.64 µm at a spatial resolution of 0.05 °× 0.05 °. In this study AOD retrieved from NOAA-10 (1987-1991), NOAA-12 (1992-1998), NOAA-15 (1992-2002), NOAA-17 (2003-2009), and METOP-ATETOP-A (2010-2014) is used. The AVHRR data from 1983 to 1986 are not used because there are Jan to May in 1983, May to Jul in 1984, Jul to Oct in 1985, and Nov to Dec in 1986 are available, not consistent.

2.2.2 ATSR (ATSR-2 and AATSR)

Two Along-Track Scanning Radiometers (together referred to as ATSR) are used in this study: the ATSR-2, which flew on ESA's ERS-2 from 1995-2003, and the Advanced ATSR (AATSR), which flew on ESA's environmental satellite ENVISAT and provided data from May 2002 until April 2012. Both satellites flew in a sun-synchronous descending orbit with a day-time equator crossing time of 10:30 LT (ERS-2) and 10:00 LT (ENVISAT). Together these instruments provided 17 years of global aerosol data. The ATSR sensor has two views (near-nadir and 55° forward from nadir) which facilitate effective separation of the surface and atmospheric contributions to the reflectance at TOA. Multiple wavelengths (7) from VIS to TIR facilitate effective cloud screening and allow for multi-wavelength retrieval of aerosol properties. ATSR has a conical scan mechanism with a swath of 512 km, resulting in daily global coverage in 5-6 days.

The ATSR dual view was first applied for AOD retrieval over land by Veefkind et al. (1998), based on the principles outlined by Flowerdew and Haigh (1995). Over ocean the two views are used separately to retrieve the AOD in both the nadir and forward directions. The ATSR dual view algorithm (ADV) has been much improved during algorithm experiments (Holzer-Popp et al., 2013) as part of the ESA Climate Change Initiative (cci) (Hollmann et al., 2013) project Aerosol_cci (de Leeuw et al., 2015, Popp et al., 2016). The most recent version of the ATSR dual view algorithm ADV is described in Kolmonen et al. (2016). ATSR L1 gridded brightness temperature data are provided with a nominal resolution of 1x1 km² sub-nadir and aerosol data are provided at a default spatial resolution of 10×10 km² on a sinusoidal grid (L2) and at 1° x 1° (L3).

The ATSR product used in this paper is the AOD at a wavelength of 550 nm, over the study area for the full ATSR mission. The data were produced using ADV version 2.31 which includes cloud post-processing as described in Sogacheva et al. (2017). ATSR-2 retrieved AOD data are available for the period June 1995- December 2003, with some gaps in 1995 and 1996. AATSR data are available for the period May 2002 - April 2012, but some data are missing in 2002 (see de Leeuw et al., 2018, for more details).

2.2.3 **MODIS**

The MODerate resolution Imaging Spectroradiometer (MODIS) flies aboard the NASA Terra and Aqua satellites, launched in December 1999 and May 2002, respectively, in a near-polar sun-synchronous circular orbit with an equator crossing time of 10:30 and 13:30 (LT), respectively (Salomonson et al., 1989). MODIS is a single view instrument with a swath of 2330 km (cross track) and provides near-global coverage on a daily basis. One of the most successful products of MODIS, which has been used in numerous aerosol related studies, is the AOD at 550 nm.

MODIS AOD is retrieved using two separate algorithms, Dark Target (DT) and Deep Blue (DB). In fact, two different DT algorithms are utilized, one for retrieval over land (vegetated and dark-soiled) surfaces (Kaufman et al., 1997, Remer et al., 2005, Levy et al., 2010, 2013) and one for retrieval over water surfaces (Tanréet al., 1997, Remer et al., 2005, Levy et al., 2013). The DB algorithm (Hsu et al., 2004, 2013) was traditionally used over bright surfaces where DT cannot be used (e.g.

deserts, arid and semi-arid areas). However, the enhanced DB algorithm is capable of returning aerosol measurements over all land types (Sayer et al., 2013, 2014). The DT Expected Error (EE) is $\pm (0.05 \pm 0.15 \tau_{AERONET})$ over land and $\pm (0.04 \pm 0.1 \tau_{AERONET})$, $\pm (0.02 \pm 0.1 \tau_{AERONET})$ over sea relative to the AERONET optical thickness ($\tau_{AERONET}$) (Levy et al., 2013). The DB expected error is $\pm (0.03 \pm 0.2 \tau_{MODIS})$ relative to the MODIS optical thickness (τ_{MODIS}) (Hsu et al., 2013, Sayer et al., 2015). In this study the recently released (end of 2017) MODIS/Terra merged C6.1 L2 ($10 \times 10 \text{ km}^2$) AOD dataset is used because of the proximity of the MODIS/Terra and AVHRR overpasses. The merged (DT and DB) dataset is described by Levy et al. (2013) and includes measurements from both algorithms, offers a better spatial coverage and can be used in quantitative scientific applications (Sayer et al., 2014). This merged data set was validated over China using AERONET data, showing the good performance for all retrieved AOD values up to 2.4, with a small overall bias of 0.06 (Sogacheva et al., 2018a).

10 2.3 Ground-based reference data

Ground-based reference data used in this study are sunphotometers un photometers from AERONET (Holben et al., 1998) and CARSNET (Che et al., 2015) available in the study area (see Fig. 1), complemented with Broadband Extinction Method (BEM)radiance derived AOD data in Beijing for the period when no sunphotometers un photometer data are available. The eight AERONET sites in the study area are all located in, or close to, Beijing. Three of them (Beijing, Beijing_RADI, and PKU_PEK) are located in the city, the others are located in rural areas in the vicinity of Beijing. The eight CARSNET sites are distributed over a wider area extending beyond BTH, with two of them (Xilinhot and Datong) located to the NW of the Heihe-Tengchong line, Huimin in the NCP and Yushe in rural areas surrounded by mountains. The other four (Lingshan_Mountain, XiangHe, Xinglong, and Yufa_PEK) are in and around Beijing. -The solar radiation station is located in Beijing, close to three AERONET sites (Beijing, Beijing_RADI, Beijing_CMA).

O The AERONET, CARSNET and radiation-derived AOD data sets are briefly described below.

2.3.1 AERONET

The AERONET project is a federation of ground-based remote sensing aerosol networks established by the National Aeronautics and Space Administration (NASA) in the USA and PHOtomérie pour le Traitement Opérationnel de Normalisation Satellitaire (PHOTONS) in France. AERONET was expanded with other networks and national efforts, see the AERONET website (https://aeronet.gsfc.nasa.gov/) for a description of contributors, sites, operational procedures, data products and availability, etc. AERONET serves as the primary network for global validation of satellite-retrieved aerosol products, including AOD which is used in this study. AOD at all AERONET stations is measured using CIMEL sunphotometersun photometers. Most common are the CIMEL CE-318 models with 5 wavelengths (440, 670, 870, 936 and 1020 nm) or polarized with 8 wavelengths, measuring direct sun as well at many angles using different scan patterns which provide the data necessary to retrieve a multitude of aerosol properties. Data checking and processing is done centrally and the products are freely available from the AERONET website. Data are made available at three levels, i.e. Level 1.0 (unscreened),

Level 1.5 (cloud-screened), and Level 2.0 (L2) (cloud-screened and quality-assured). The uncertainty of the CIMEL-derived AOD is 0.01-0.02 (wavelength dependent) (Eck et al., 1999). In this study we use AERONET L2 data from the most recent V3 dataset for validation and comparison (Giles et al., 2018). Satellite AOD data are commonly made available at 550 nm, and hence, for validation, the AERONET data are interpolated to this wavelength by using the Ångstr äm Exponent (AE) which describes the AOD wavelength dependence (Ångstr äm, 1924).

Almost all AERONET sites in the study area started observations after 2000 as Tab. 1 shows. Long-time and continuous measurements from 2001 to 2014 are only available from the Beijing and XiangHe stations. Therefore, we selected these stations for comparison with satellite time series, but for validation all available data are used.

Table 1. Data availability from AERONET sites in the study area (35 ° 45 °N, 110 ° 120 °E)

	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
Beijing	√	√	√	√	√	√	√	√	√	√	√	√	√	√
Beijing-CAMS												\checkmark	√	\checkmark
Beijing_RADI										√	√			
Lingshan_Mountain														√
XiangHe	\checkmark			√	\checkmark	√	√	√	√	√	√	√	√	√
Xinglong						√	√	\checkmark	\checkmark	√	√	√		√
PKU_PEK						√		\checkmark						
Yufa_PEK						√								

10 2.3.2 CARSNET

AERONET sites in the study area are all located in or around Beijing (cf. Fig. 1). To expand the reference data set to other regions, AOD data from CARSNET (Che et al., 2015) was used, but only data for the years 2007, 2008 and 2010 were available for this study. The locations of the CARSNET sites in the study area are indicated in Fig. 1. CARSNET was established by the China Meteorological Administration (CMA) for the study of aerosol optical properties and validation of satellite retrievals, and the same instrumentation as AERONET (i.e. CIMEL CE-318) and similar procedures (Che et al., 2015). The difference is the way of instrument calibration and AOD calculation as described in Che et al. (2009). The CARSNET AOD uncertainties are 0.03, 0.01, 0.01 and 0.01 at the 1020, 870, 670 and 440 nm channels (Che et al., 2009).

2.3.3 Radiation-derived Broadband extinction method (BEM)BEM AOD data

Broadband solar radiation has been measured in China by China Meteorological Administration (CMA) since the 1950s at 98 sites (Qiu, 1998) which evolved into the China national solar radiation network with 14 stations (Xu et al., 2015) providing

continuous data since 1993 using China-made pyrheliometers. Methods were developed to retrieve monochromatic or equivalent AOD using hourly accumulated direct solar radiation and the results of these broadband extinction methods (BEM AOD) are in good agreement with sunphotometersun photometer data (see Xu et al., 2015 for an overview). In the current study we used BEM AOD at 550 nm retrieved from the pyrheliometer measurements of broadband solar radiation at the Beijing station (Fig. 1) of the China national solar radiation network. These data are particularly useful for evaluation of the AVHRR-retrieved AOD for the period before 2000 when no sunphotometersun photometer data were available. The AOD was retrieved using the methodBEM described by Xu et al. (2015). Xu et al. (2015) evaluate the quality of the BEM-derived AOD (at 750 nm) for the Beijing station from comparison with AERONET AOD. The results show that 59% of the hourly AOD data fall within an error envelope of ±(0.05+0.15\tau_{AERONET}). For monthly averaged data this is 82%. Guo et al (2016) used the hourly mean BEM-derived hourly mean AOD at 550 nm to evaluate the products from MODIS, OMI and MISR, obtaining consistent results with previous validations based on sunphotometersun photometer measurements, which proved the effectiveness of the radiation-derivedBEM AOD in satellite product validation.

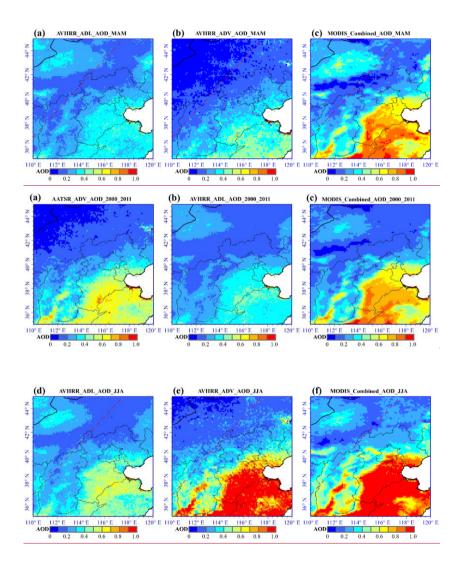
3 Result

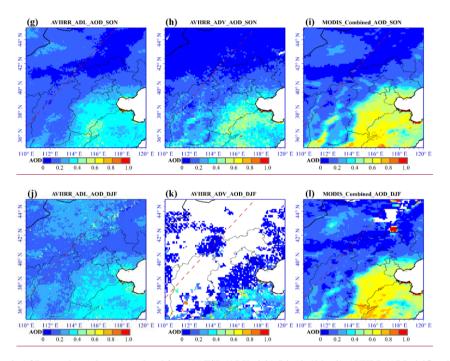
5 3.1 Data overview

The aim of the present study is to determine the compatibility of AVHRR and ATSR AOD data sets and their suitability to combine them to fill the gap between AATSR and SLSTR, as well as to extend the ATSR time series to the 1980s. AVHRR AOD data is available over the study area from the ADL algorithm for the years 1983-2014 (Xue et al., 2017) and from ATSR-2 and AATSR using the ADV algorithm for the years 1995-2003 (ATSR-2) and 2002-2012 (AATSR), with some gaps as described in de Leeuw et al. (2018). The consistency between ATSR-2 and AATSR AOD is addressed in Sogacheva et al. (2018a) who show that no systematic differences occur over China. In the current work we also use MODIS/Terra C6.1 merged DTDB AOD for comparison. The validation of this data set over China by Sogacheva et al. (2018a), using all available AERONET stations, shows it's good quality, with a small positive bias (0.06) with respect to AERONET AOD. As an example, AOD maps over the study area are shown in Fig. 2, seasonal aggregated for the full years 2000-2011 when data from all three sensors/algorithms are available: AATSR (ADV v2.31, L2, resolution 10x10 km²), AVHRR (ADL, resolution 0.05 °x 0.05 °9 and MODIS C6.1 Merged DBDT (L2, resolution 10x10 km²).

Figure 2 shows the similar AOD patterns derived from the three sensors/algorithms, with high AOD in the SE part of the study area and lower elsewhere. As indicated above, these high and low AOD areas are roughly separated by the Heihe-Tengchong line which separates the NCP from the Loess Plateau in Shanxi Province and the Inner Mongolia Plateau. In the SW of the study area, in Shanxi Province, we see an area with elevated AOD stretching from the NE to the SW, i.e. in the Guanzhong

basin where pollution transported by NE winds from the BTH area accumulates between the Qinling Mountains and the Loess Plateau and mixes with locally produced pollution. Comparison of the spatial distribution shows the similarity of the AOD distributions for AVHRR and MODIS while for AATSR the spatial distribution deviates with low AOD over Inner Mongolia, i.e. over the Gobi desert with high surface reflectance. ADV AOD retrieval is known to often be unsuccessful over bright areas and hence also aggregated AODs are too low (de Leeuw et al., 2018, Sogacheva et al., 2108a). AVHRR ADL appears to produce more credible AOD values in such conditions, as suggested by the similarity of the AOD patterns to those from MODIS. Below this statement will be put in perspective with the validation results from the ground-based reference data set. Quantitatively, Figure 2 shows that in the south of the study area, i.e. south of 41 N, the MODIS AOD is overall higher than that retrieved from AATSR which in turn is overall somewhat higher than that from AVHRR. The smoother MODIS AOD, likely due to the larger number of data points because of the larger swath, together with the higher AOD and the scales chosen to plot the AOD maps, results in larger variability and patterns which are not as clearly revealed by ATSR and AVHRR. North from 41° AATSR AOD retrievals are often not successful so a quantitative comparison cannot be made at these latitudes. As regards the comparison of the other two sensors north of 41 N MODIS is overall higher than AVHRR, except, for instance, over an area in Inner Mongolia just north of 41 N where the MODIS AOD is close to zero while AVHRR provides AOD values between 0.1 and 0.2.





 $Figure 2: AOD over the study area retrieved from AATSR (ADV v2.31, L2, 10x10 km2), AVHRR (ADL, 0.05 ° x 0.05 °) and MODIS C6.1 Merged DTDB (MODIS C6.1, L2, 10x10 km2), <math display="block">\underline{seasonal} \ aggregated over the years 2000-2011.$

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Below the satellite AOD data will be validated versus the ground-based reference data from AERONET and CARSNET, for all stations available in the study area, as indicated above. The AVHRR data will also be compared with the radiation-derived AOD data available from the Beijing station which are especially useful for the earlier period when no other reference data are available. Next the satellite-derive AOD time series will be compared with the reference data to evaluate for each of the data sets when they are most useful. For AVHRR and MODIS, with wide swath, this will be done using monthly averaged AOD. For ATSR, with a much smaller swath, the data volume is too small for monthly averaging in a statistically meaningful sense and therefore seasonal averages will be used. In the direct comparison of monthly time series from other sensors, this may lead to an apparent shift in the AOD peak values.

3.2 Data quality: validation

3.2.1 Procedure

The MODIS and ATSR AOD data sets used in this study have been validated versus sunphotometersun photometer reference data, on global and regional scales. With a focus on China, the ATSR v2.31 and MODIS C6.1 have been validated using AERONET data, for mainland China, for ten regions across China and for different seasons (de Leeuw et al., 2018, Sogacheva et al., 2018a). However, as shown in de Leeuw et al. (2018) and Sogacheva et al. (2018a), strong regional differences occur in both the seasonal and long-term AOD variations. Therefore, to achieve an unbiased comparison between the different data sets over the current study area, the MODIS C6.1 and ATSR data used in this study are validated versus the available reference data, together with the AVHRR data. The ADL data over the study area was earlier validated by Xue et al. (2017), but only AERONET stations were used while in the current study the complementary information (AOD) from CARSNET is also used.

Collocation of satellite and reference data is important for validation. Here we follow the spatio-temporal collocation method proposed by Ichoku et al., (2002), i.e. the satellite data were averaged over an area of 5 x 5 pixels (ca. 50 x 50km² at nadir) around the sunphotometersun photometer location, whereas the sunphotometersun photometer data were averaged over ±30 min around satellite overpass time. This spatio-temporal collocation method has been widely used for the validation of satellite aerosol products, for instance for MODIS (Ichoku et al., 2002, Chu et al., 2002, Remer et al., 2005, Levy et al., 2010, Sayer et al., 2014), AATSR (Che et al., 2016), and AVHRR (Riffler et al., 2010, Xue et al., 2017).

In the data presentation and discussion expected error (EE) envelopes are used which apply to MODIS \underline{DT} over land, i.e. expected errors of $\pm (0.05\pm 0.15\tau_{AERONET})$ (Levy et al., 2013). For comparison, this value is also used for ATSR and AVHRR although the actual expected error envelopes for these sensors, which were not designed for aerosol retrieval, is expected to be higher than for MODIS. For ATSR a per-pixel uncertainty is provided rather than EE (Kolmonen et al., 2016) and for the AVHRR ADL (Xue et al., 2017) uncertainties have not been estimated.

The Beijing solar radiation station is located to the SW of the Beijing AERONET site at a distance of approximately 20km. AOD differences at these two sites are likely to occur due to emissions and secondary formation processes over this urban area, but these have not been accounted for in the current study where collocations include averages over areas larger than the separation between these sites. Therefore, radiation retrieved BEM AOD is directly compared to the sunphotometersun photometer AOD as was done by Xu et al. (2015).

3.2.2 Results

Figure 3 shows scatterplots of AOD, for each of the three data sets individually, versus ground-based AOD reference data from all AERONET and CARSNET (3 years: 2007, 2008 and 2010) sites in the study area. Large differences occur between the three data sets. Starting with MODIS/Terra C6.1 merged DTDB, which has the largest number of collocations, the scatterplot in Fig. 3a shows the excellent performance for AOD up to 1.3 with the bin-averaged AOD less than 0.05 below the

identity line and 69.5% of all data points within expected error of $\pm (0.05 \pm 0.15\tau_{AERONET})$. For AOD of 1.3 and larger the bin-averaged AOD values are much lower than the reference values, although still within one standard deviation. For AOD>2.6, MODIS does provide values but they are all well below the identity line. These results indicate that MODIS C6.1 AOD over the study area is reliable for AOD up to 1.3, but for higher AOD their use is not recommended.

- ATSR with a swath width of 500 km provides much less collocations than MODIS but the large difference in performance (compare Fig. 3a and 3c) cannot be explained by the swath alone and is likely due to the failure of ADV to provide a retrieval over bright surfaces and the underperformance in the winter with dry conditions, low humidity, little precipitation, low light levels and limited or no plant growth resulting in relatively high surface reflectanceadequate retrievals over bright surfaces. Figure 3b shows that ADV provides AOD values up to 2.7 but for AOD > 1.3 the values are widely scattered around the identity line with large deviations from the sunphotometersun photometer values. Also for lower AOD a systematic underestimation is observed which increases with the AOD value up to 0.8 where the ADV-retrieved AOD is about 0.3 low. For larger AOD, up to 1.3, ADV continues to underestimate the AOD with a similar amount. Although 52% of the data points are within the EE, ADV clearly underestimates the AOD over this area and the bin-averaged AOD follows the lower EE limit rather than the identity line, for AOD up to 1.0, leading to the conclusion that ADV AOD is about 0.15 x AOD low.
- AVHRR with a swath width of 2900 km is expected to provide the largest number of collocations but actually it is about 20% lower than for MODIS, likely due to failure of ADL to provide a valid retrieval in all situations. ADL does not provide retrievals for AERONET AOD > 2 and the data in Fig. 3a show the large scatter around the identity line and the large underestimation for AOD > 0.5. For AOD larger than about 0.8, the binned ADL AOD data deviate from the identity line by more than the expected error. For AOD up to 0.5, the bin-averaged values are within the EE (58.1% of the data), but systematically deviate from the identity line and underestimation increases with increasing AOD.
 - In conclusion, in the study area, MODIS provides reliable AOD for values up to 1.3 with a slight underestimation. This is in contrast with the findings of Sogacheva et al. (2018a) who observed a slight overestimation (bias 0.06) of the MODIS/Terra C6.1 merged DTDB AOD using all AERONET data available over China, but no CARSNET. CARSNET could provide more reference data where there are not AERONET data except Beijing region. Also for ATSR ADV v2.31 data the validation presented in de Leeuw et al. (2018), also using only AERONET data similar to Sogacheva et al. (2018a), deviates from that presented here, with a slight underestimation resulting in a bias of 0.07. Clearly the selection of the smaller study area affects the validation results, with for MODIS slightly better results (smaller bias) than over all China, but for ATSR ADV the performance is less good. AVHRR AOD shows a large scatter and does not follow the identity line even for low values, underestimates for AOD larger than 0.5 and appears to fail for AOD larger than about 0.8.

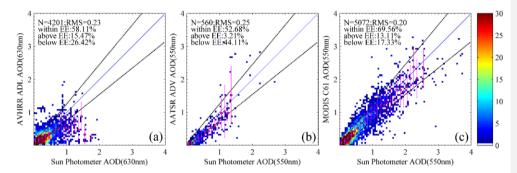


Figure 3: Scatter density plots of satellite-retrieved AOD versus sunphotometers photometer retrieved AOD data from AERONET (all available data) and CARSNET (3 years: 2007, 2008 and 2010) stations in the study area. The red symbols represent the averaged satellite-retrieved AOD binned in 0.1 sunphotometers photometer AOD intervals and the vertical lines on each circle represent the 1-sigma standard deviation of the fits. The colour bar on the right indicates the number of data points. Parameters for a LSQ fit to all data points are presented in the upper left corner, where N is the total number of collocated pairs and RMS is the root mean square error. The blue line represents the identity line and the black dotted lines represent the MODIS EE.

Table2. Metrics obtained from the evaluation of three AOD products. N is number of collocated data points, MSA is mean satellite retrieved AOD, MAA is mean sunphotometersun photometer (AERONET and CARSNET) derived AOD, RMB is the ratio MAA/MSA. MBE is mean of AOD bias given by $\tau_{sat} - \tau_{aero}$. R is the Pearson correlation coefficient for a LSQ fit to all data points. RMS is the root mean square error. EE is the expected error, EE_a is the fraction above the EE, EE_b is the fraction below the EE.

Product	Network	N	MSA	MAA	RMB	MBE	RMS	R	EE	EE_a	EE_b
MODIS	AERO&CARS	5072	0.441	0.464	0.950	-0.023	0.204	0.910	69.56%	13%.11	17.33%
ADV	AERO&CARS	560	0.289	0.396	0.729	-0.107	0.252	0.843	52.68%	3.21%	44.11%
AVHRR	AERO&CARS	4201	0.237	0.296	0.802	-0.059	0.229	0.602	58.11%	15.47%	26.42%

Seasonal scatterplots are presented in the Appendix and discussed in the context of the seasonal variations observed in the AOD time series presented below.

3.3 Comparison of satellite-derived AOD time series with reference data

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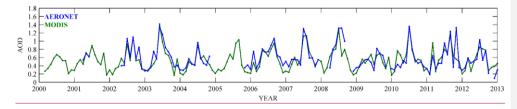
In this section time series of satellite-retrieved AOD data will be compared with time series available from the AERONET sites in Beijing and XiangHe, for which the longest time series are available, and with AOD derived from broadband radiances measured at the Beijing radiation station.

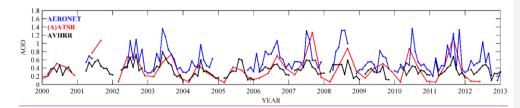
3.3.1 AERONET AOD in Beijing

AOD time series for the period 19972000-20125, retrieved from AVHRR, ATSR and MODIS data and spatially collocated with the Beijing AERONET site, are presented in Fig. 4 together with monthly-averaged AERONET AOD for reference. The time series presented here start in 2000 because for the earlier years no AERONET and MODIS data are available. The data in Fig. 4 show that AVHRR is almost always lower than AERONET and that the high AOD values obtained from AERONET, usually in the summer, are not reproduced by AVHRR. This is not unexpected since the scatterplots in Fig. 3 show that AVHRR fails to retrieve high AOD. For these individual data points hardly any values larger than 1 were retrieved and hence the monthly averages in Fig. 4 are lower than that. Figure 4 shows that the highest AVHRR AOD values of about 0.8 occur in 2002 and 2003 and thereafter do not exceed 0.6. The monthly averaged AERONET AOD peak values occur mostly in the summer and are about 1.4. However, in other seasons the differences are much smaller and the two time series seem to trace rather well with an offset of about 0.1. In particular in the winter AVHRR AOD is close to the reference value. These seasonal differences are confirmed by the scatterplots presented in the Appendix. The results lead to the conclusion that, for the Beijing site, for low AOD the ADL algorithm provides quite reasonable results but for high AOD improvement is needed to provide reliable time series.

The MODIS data in Fig. 4 (top) trace the AERONET AOD very well, both in the summer and in other seasons, with MODIS AOD often just a little smaller as can also be observed from the scatterplots in the Appendix. This shows that MODIS serves as a good reference for the situation encountered at the Beijing site.

This does not apply to the ATSR AOD. Although ATSR AOD variations follow those from AERONET, and even reproduce the high summer values in some years (2007 and 2011), the ATSR values are most of the time smaller and especially in the winter the ATSR AOD is much too low. As discussed before, this may be due to the failure of ADV to produce a valid retrieval over bright surfaces.





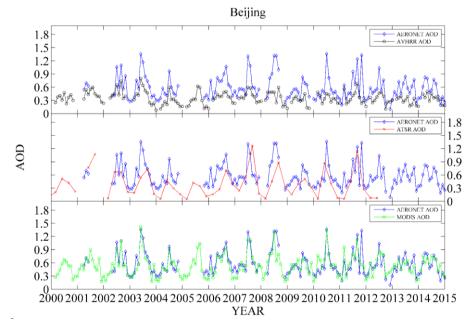


Figure 4: AOD time series for MODISAVHRR (top), (A)ATSR (middle) and MODIS AVHRR (bottom), with AERONET AOD (blue) as reference, over the Beijing site for the years 2000-20125. For (A)ATSR seasonally averaged AOD is plotted, for AVHRR and MODIS the AOD data are monthly averages.

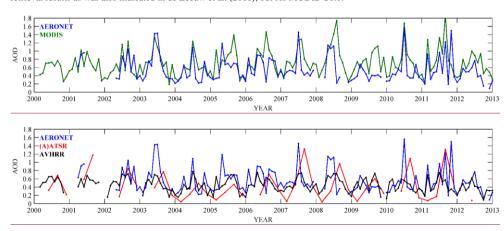
3.3.2 AERONET AOD in XiangHe

Figure 5 shows a comparison of the AOD time series over the XiangHe AERONET site, similar to that over Beijing in Fig. 4. It is noted that, for comparison, in both cases the time series are plotted for the years 2000-20125, but that the XiangHe time series start at the end of 2004 (except for a few data points in 2001), i.e. later than in Beijing. Comparison of the AERONET time series shows that over XiangHe the AOD in the summer is somewhat higher and peaks often in the same years as over

Beijing, but there are also differences such as in 2008 when the AOD in XiangHe reached the maximum monthly-averaged value of 1.5. As in Beijing, the AVHRR retrieval did not reproduce these high values in the summer, but for the other months the AVHRR/AERONET comparison is better than in Beijing and, with some exceptions, the AOD data trace very well. The seasonal scatter density plots in the Appendix, Fig. A3, confirm that no high AOD is retrieved in the summer, as opposed to other seasons. These scatterplots also confirm the underestimation of ADL, by about one EE, in each of the seasons except in the winter where ADL performance compares better to AERONET.

In contrast, the ATSR retrieval algorithm does reproduce the higher values, and in some years the seasonally averaged AOD compares favourably with the AERONET AOD (e.g. in 2007 and 2011), but in other years, such as 2008 and 2010 the maximum AOD is not observed by AATSR, which in part may be due to the seasonal averaging. Also, as over Beijing, ATSR retrieval does not work well in the winter time, possibly due to the high surface reflectance in the dry season, and the seasonal AOD is much lower than AERONET.

For MODIS the AOD compares well with AERONET but not as good as over Beijing. During some periods, e.g. in 2005 and during 2008-2011, MODIS is too high. This is also shown by the validation results in the Appendix. The reason for this difference between Beijing and XiangHe, which is located close to Beijing to the ESE (see Fig. 1), has not been further investigated. Possibly, the surface properties, urban and build-up for Beijing versus cropland for XiangHe, may affect the retrieval results as was also indicated in de Leeuw et al. (2018), but for MODIS C6.0.



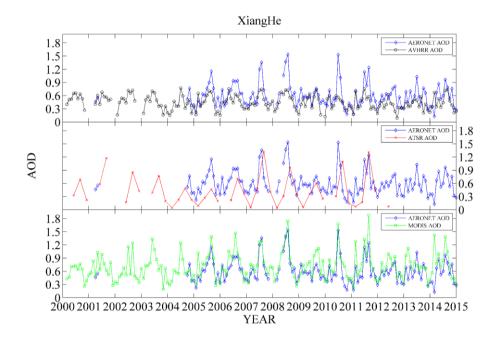


Figure 5: AOD time series for AVHRR (top), ATSR (middle) and MODIS (bottom), with AERONET AOD (blue) as reference, over the XiangHe site for the years 2000-2015. For ATSR seasonally averaged AOD is plotted, for AVHRR and MODIS the AOD data are monthly averages. AOD time series for MODIS (top), (A)ATSR and MODISAVHRR (bottom), with AERONET AOD (blue) as reference, over the XiangHe site for the years 2000-2012. For ATSR seasonally averaged AOD is plotted, for AVHRR and MODIS the AOD data are monthly averages.

3.3.3 Radiance-derived Broadband extinction method (BEM)BEM AOD in Beijing

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Figure 6 shows the comparison of the monthly-averaged AOD time series from AVHRR with those derived from radiance BEM data, as well as a comparison of the radiance BEM AOD with AERONET and with MODIS, as reference data. Monthly radiance data at the Beijing site have been validated by Xu et al. (2015) for the years 2002-2012 using both MODIS C5 L3 AOD and AERONET L2 data (version 2). In the current study MODIS C6.1 and AERONET version 3 are used, for the years 2000-2012, and the results from the comparison of the BEM radiance derived AOD with these data sets, shown in Fig. 7 (left), are similar to those presented in Fig. 2 of Xu et al. (2015). Also the time series in Fig. 6 show very good comparisons between the radiance derived BEM and MODIS and AERONET AOD. Hence the radiance derived BEM AOD data provide another reference data set for the AVHRR-retrieved AOD which can be used for the period 1987-20002012 as presented in Fig. 8.

Figure 8 shows that in this earlier period (1987-2000) the AOD was generally lower and the monthly averaged AOD compares better with BEM-radiance AOD than for the later period 2000-2012, when AOD was higher during especially the summer months. This is consistent with in right scatter in fig. 7 that almost all of AVHRR-BEM matchups are below identify line when AOD larger than 0.6. The overall conclusion remains the same, however, that the AVHRR retrieval needs improvement for high AOD and only values up to about 0.6 can be used.

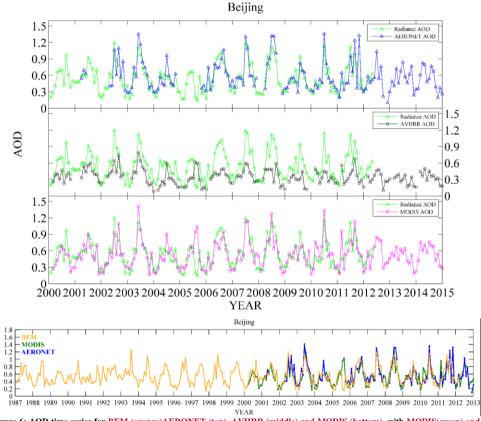


Figure 6: AOD time series for <u>BEM (orange)AERONET (top)</u>, <u>AVHRR (middle) and MODIS (bottom)</u>, with <u>MODIS(green) and</u> AERONET AOD (blue) as reference, over the Beijing site for the years <u>19872000</u>-201<u>2</u>5.

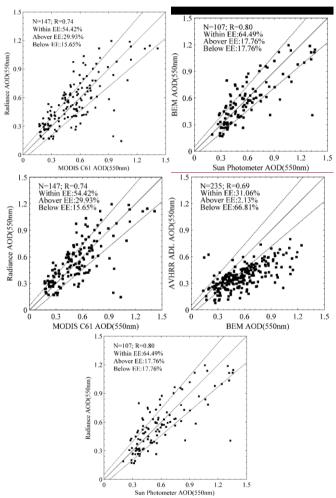


Figure 7: Scatter plots of radiance retrieved BEM AOD versus MODIS AOD, and suphotometers un photometer AOD, and AVHRR AOD. The dashed lines show the MODIS expected error of ±(0.05+0.15 targoner).

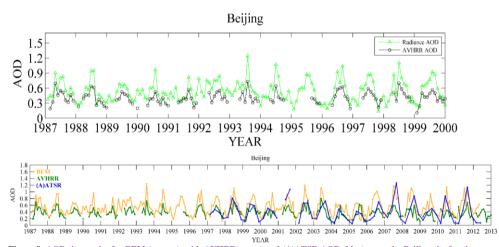


Figure 8: AOD time series for BEM (orange), with AVHRR(green) and (A)ATSR AOD (blue), over the Beijing site for the years 1987-2012Comparison of BEM radiance-derived and AVHRR-retrieved AOD time series over the Beijing site for the years 1987-20122000.

3.3 Seasonal comparison of satellite-derived AOD

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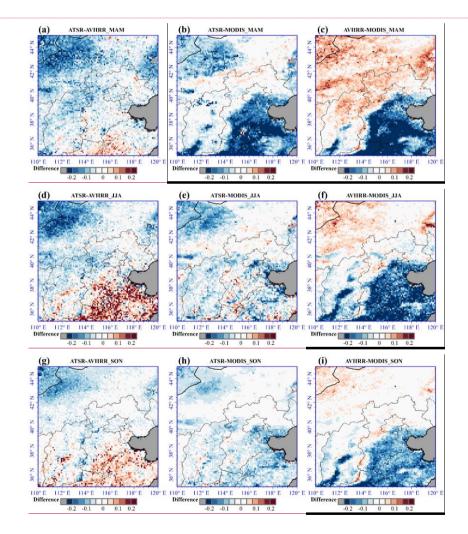
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The above results and those from Sogacheva (2018a) and de Lueeuw (2018) show MODIS data are with high quality over China. Therefore, we select MODIS as reference data both on time series and spatial coverage. Last session has compared satellite-retrieved data with ground-based data on time series, this session will compare ATSR and AVHRR data with MODIS data spatially.

As Fig. 2 shows, MODIS could provide AOD higher than 0.7 in SE part where ATSR and AVHRR data are with close pattern that hardly AOD higher than 0.4. Hence, in spring the SE part in Fig. 9b&c present same AOD difference that lower than MODIS more 0.2 and Fig. 9a present no obvious difference which is AOD difference of ATSR minus AVHRR. When the season is changed to summer time, AOD in SE part is increasing higher than 0.8 even 1.0, ATSR and MODIS could capture high AOD well but AVHRR cannot. This leads Fig. 9f presents dark blue in SE part. The situation in autumn is quite like it in spring when AOD in SE part is decreasing lower than 0.7, the patterns of ATSR and AVHRR are quite similar in SE which leads AOD difference in Fig. 9g&h are quite similar. Due to ability of ADV in winter, ATSR miss retrievals approximately in half of areas but remain part are close to AVHRR and MODIS as Fig. 9j&k show.

As discussed above, there are not enough successful retrievals at north of 41°, so we don't discuss AOD difference over part at north from 41°, otherwise AVHRR follows MODIS well in this part with slight overestimation lower than 0.1. This mainly because AVHRR has longer spectral band leading to less sensitivity to low AOD. In addition, ATSR data sample volume is much lower than other two due to sensor swath and algorithm ability, so seasonal averaged AOD are not smooth as other two.

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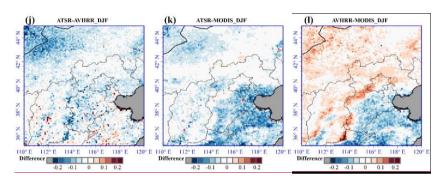


Figure 9: AOD difference over the study area retrieved from AATSR (ADV v2.31, L2, 10x10 km²), AVHRR (ADL, 0.05 °x 0.05°) and MODIS C6.1 Merged DTDB (MODIS C6.1, L2, 10x10 km²), seasonal aggregated over the years 2000-2011.

5 4 Discussion and conclusion

The validation exercises and time series comparisons presented in Sect. 3 show the stronger and weaker points of the ATSR-and AVHRR-retrieved AOD Climate Data Records (CDRs), with AVHRR being better in the winter and ATSR better in the summer. These differences are also very clear in the direct comparison of the AVHRR and ATSR time series, shown in Fig. 109 for the Beijing site and in Fig. 10-11 for the XiangHe site. Figure 910 also shows that before 2007 these time series trace quite well and could be combined into a single time series while taking into account the validation results for that period only. In particular the use of the ATSR winter AOD data will have to be considered carefully and may possible have to be given little weight. However, after 2007 large differences are observed with quite high ATSR AOD in the summer while that retrieved from AVHRR has a maximum value of about 0.6. The comparison with MODIS shows that the ATSR AOD values are credible and follow those of MODIS (apart from the shift due the use of seasonal ATSR vs monthly MODIS AOD averages), whereas the AVHRR AOD is much too low. In the winter the opposite is observed: ATSR is too low with values close to zero while AVHRR traces MODIS very well.

The intercomparison of the ATSR and AVHRR time series for XiangHe in Fig. 10-11 shows a similar pattern as for Beijing, with a good comparison before 2007 and a larger summer/winter difference in the ATSR data there-after, resulting in increased discrepancy between the ATSR and AVHRR data sets. However, the comparison with MODIS shows an offset between ATSR and MODIS in the years 2004-2007 which seems to be due to larger overestimation of the MODIS AOD at the XiangHe site than at the Beijing site. Also in some other years the ATSR/MODIS difference appears to be higher in XiangHe than in Beijing, due to higher MODIS AOD. The comparison between AERONET and MODIS AOD data in Figures 4 and 5 show that the

difference between the AOD measured at both sites is smaller for AERONET than that for MODIS and hence this may be a MODIS retrieval issue.

As the statistics marked in A2 and A3, the data volume of collocated matchups with ground-based reference data may lead to biased validation results and time series comparison. Therefore, pixel by pixel comparison with reference data is necessary. The seasonal averaged AOD and AOD difference maps provides information on not only seasonal but also pixel by pixel comparison. As reference data, MODIS AOD could be not only sensitive to low AOD but also used to detect high AOD that it performs best in different seasons (see A1). As discussed above, AVHRR failed to retrieve AOD over 0.6 for most cases leading to big differences with ATSR and MODIS in the summer in the SE (of study area). However, the AOD patterns of ATSR and AVHRR in other seasons are similar. ATSR has ability to retrieve high AOD, otherwise its data coverage limits its performance as Fig. 9 and c shows that difference between ATSR and MODIS AOD is close to zero while the difference

between AVHRR and MODIS AOD is lower than -0.2 for most of area of the SE. This is consistent with last conclusion.

BEM AOD data set could not only provide consistent AOD from 1980s to the current but also be with credible data quality compared with MODIS and AERONET AOD. As shown in Fig. 8, BEM AOD agrees MODIS and AERONET AOD very well with close pattern at overlaying years. Above, it is concluded that AVHRR AOD agrees ATSR well, however there is no ATSR AOD when it goes back to early 1990s and 1980s, AVHRR is the only choice to extend ATSR AOD data set. Still, AVHRR AOD missed most of high AOD over 0.6 in the summer but follows BEM AOD well in other seasons and has similar annual patterns. Only one BEM station weaker conclusion of this paper, however there are at least ten BEM stations in whole China region as mentions in Xu et al. (2015), hence it is expected to extend AVHRR data set to whole China (which is our current focus).

In conclusion, the possible combination of the AVHRR and ATSR CDRs, possibly also including MODIS, into a multi-decadal time series is not straightforward. Although MODIS performance is better that that of ATSR and AVHRR, the latter two sensors go back in time before MODIS and hence provide a unique source of information. In addition, none of the satellite sensors performs better than the others at any time and at any location and their combination taking into account the strengths and weaknesses of each of them, may result in a more significant CDR than each of them individually as indicated by that AVHRR failed to retrieve AOD over 0.6 while ATSR could and ATSR AOD is not credible from 41° to the north border while AVHRR AOD is acceptable. Furthermore, the comparison shows that before 2007 the AVHRR and ATSR compare quite well and could be used for a meaningful extension of the AOD data records to the early 1980s. However, this conclusion does not apply to the northern sub-region where the surface conditions seem unfavourable for AOD retrieval from ATSR using ADV.

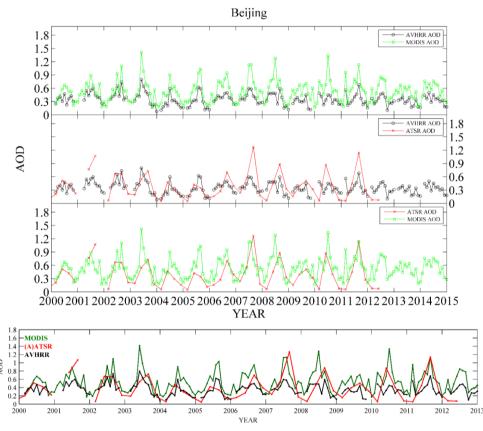


Figure 109: Comparison of AOD time series over the Beijing AERONET site retrieved from the satellite sensors AVHRR, MODIS and (A)ATSR, for the overlapping years 2000-2015. Note that for AVHRR and MODIS monthly mean AOD data are plotted, while for (A)ATSR they are seasonal means which may provide a shift in the extreme values.

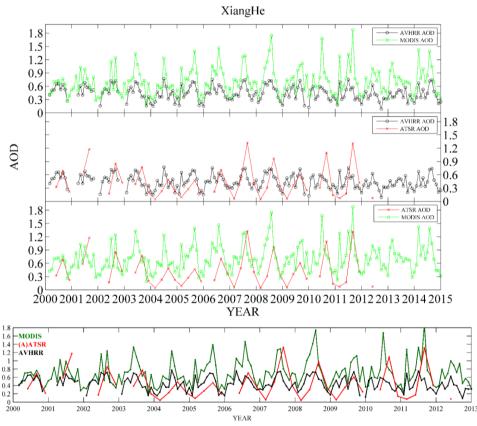


Figure 1011: As Fig. 9101, over the XiangHe site.

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5 References

10

15

- Allen, R.C., Durkee, P.A., and Wash, C.H.: Snow/Cloud Discrimination with Multispectral Satellite Measurements, J. Appl. Meteorol. Clim., 29, 994-1004, doi: 10.1175/1520-0450(1990)029<0994:SDWMSM>2.0.CO;2, 1990.
- Alizadeh-Choobari, O., Sturman, A., and Zawar-Reza, P.: A global satellite view of the seasonal distribution of mineral dust and its correlation with atmospheric circulation, Dynam. Atmos. Oceans, 68, 20–34, doi: 10.1016/j.dynatmoce.2014.07.002, 2018.
- Ångström, A.: Solar and terrestrial radiation. Report to the international commission for solar research on actinometric investigations of solar and atmospheric radiation, Q. J. Roy. Meteor. Soc., 50, 121-126, doi:10.1002/qj.49705021008, 1924.
- Bai, J., de Leeuw, G., van der A, R., De Smedt, I., Theys, N., Van Roozendael, M., Sogacheva, L., and Chaia, W.: Variations and photochemical transformations of atmospheric constituents in North China, Atmos. Environ., 189, 213-226, https://doi.org/10.1016/j.atmosenv.2018.07.004, 2018.
- Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380–5552, doi: 10.1002/jgrd.50171, 2013.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann,
 U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B., and Zhang, X.: Clouds and Aerosols. In: Climate Change 2013:
 The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental
 Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.
- Brimblecombe, P.: The Clean Air Act after 50 years, Weather, 61, 311–314, doi: 10.1256/wea.127.06, 2006.
- Carn, S. A., Fioletov, V. E., McLinden, C. A., Li, C., and Krotkov, N. A.: A decade of global volcanic SO2 emissions measured from space, Sci. Rep., 7, 44095, doi: 10.1038/srep44095, 2017.
- Che, H., Zhang, X. Y., Chen, H. B., Damiri, B., Goloub, P., Li, Z. Q., Zhang, X. C., Wei, Y., Zhou, H. G., Dong, F., Li, D. P., and Zhou, T. M.: Instrument calibration and aerosol optical depth validation of the China Aerosol Remote Sensing Network, J. Geophys. Res., 114, D03206, doi:10.1029/2008JD011030, 2009.

- Che, H., Zhang, X.-Y., Xia, X., Goloub, P., Holben, B., Zhao, H., Wang, Y., Zhang, X.-C., Wang, H., Blarel, L., Damiri, B., Zhang, R., Deng, X., Ma, Y., Wang, T., Geng, F., Qi, B., Zhu, J., Yu, J., Chen, Q., and Shi, G.: Ground-based aerosol climatology of China: aerosol optical depths from the China Aerosol Remote Sensing Network (CARSNET) 2002–2013, Atmos. Chem. Phys., 15, 7619–7652, doi: 10.5194/acpd-15-12715-2015, 2015.
- 5 Che, Y., Xue, Y., Mei, L., Guang, J., She, L., Guo, J., Hu, Y., Xu, H., He, X., Di, A., and Fan, C.: Technical note: Intercomparison of three AATSR Level 2 (L2) AOD products over China, Atmos. Chem. Phys., 16, 9655–9674, doi: 10.5194/acp-16-9655-2016, 2016.
 - Chen, J., Li, C., Ristovski, Z., Milic, A., Gu, Y., Islam, M. S., Wang, S., Hao, J., Zhang, H., He, C., Guo, H., Fu, H., Miljevic, B., Morawska, L., Thai, P., LAM, Y. F., Pereira, G., Ding, A., Huang, X., Dumka, U. C.: A review of biomass burning: Emissions and impacts on air quality, health and climate in China, Sci. Total Environ., 579, 1000–1034, doi: 10.1016/j.scitotenv.2016.11.025, 2017.

15

- Chu, D. A., Kaufman, Y. J., Ichoku, C., Remer, L. A., Tanre , D., and Holben, B. N.: Validation of MODIS aerosol optical depth retrieval over land, dust, Geophys. Res. Lett., 29, MOD2-1–MOD2-4, doi: 10.1029/2001GL013205, 2002.
- de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.: Production flux of sea spray aerosol, Rev. Geophys., 49, doi: 10.1029/2010RG000349, 2011.
- de Leeuw, G., Holzer-Popp, T., Bevan, S., Davies, W. H., Descloitres, J., Grainger, R. G., Griesfeller, J., Heckel, A., Kinne, S., Klüser, L., Kolmonen, P., Litvinov, P., Martynenko, D., North, P., Ovigneur, B., Pascal, N., Poulsen, C., Ramon, D., Schulz, M., Siddans, R., Sogacheva, L., Tanré D., Thomas, G. E., Virtanen, T. H., von Hoyningen-Huene, W., Vountas, M., and Pinnock, S.: Evaluation of seven European aerosol optical depth retrieval algorithms for climate analysis, Remote Sens. Environ., 162, 295-315, doi:10.1016/j.rse.2013.04.023, 2015.
- de Leeuw, G., Sogacheva, L., Rodriguez, E., Kourtidis, K., Georgoulias, A. K., Alexandri, G., Amiridis, V., Proestakis, E., Marinou, E., Xue, Y., van der A, R.: Two decades of satellite observations of AOD over mainland China using ATSR-2, AATSR and MODIS/Terra: data set evaluation and large-scale patterns, Atmos. Chem. Phys., 18, 1573-1592, doi: 10.5194/acp-18-1573-2018, 2018.
- 25 Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, J. Geophys. Res.-Atmos., 104, 31333–31349, doi: 10.1029/1999JD900923, 1999.
 - Flowerdew, R. J., and Haigh, J. D.: An approximation to improve accuracy in the derivation of surface reflectance from multi-look satellite radiometers, Geophys. Res. Lett., 23, 1693–1696, doi: 10.1029/95GL01662, 1995.
- 30 Giles, David M., Sinyuk, Alexander, Sorokin, Mikhail G., Schafer, Joel S., Smirnov, Alexander, Slutsker, Ilya, Eck, Thomas F., Holben, Brent N., Lewis, Jasper R., Campbell, James R., Welton, Ellsworth J., Korkin, Sergey V., Lyapustin, Alexei I.: Advancements in the Aerosol Robotic Network (AERONET) Version 3 database automated near-real-time quality control algorithm with improved cloud screening for Sun photometer aerosol optical depth (AOD) measurements, Atmos. Meas. Tech., 12, 169–209, doi: 10.5194/amt-12-169-2019, 2019.

- Fioletov, V. E., McLinden, C. A., Krotkov, N., Li, C., Joiner, J., Theys, N., Carn, S., and Moran, M. D.: A global catalogue of large SO2 sources and emissions derived from the Ozone Monitoring Instrument, Atmos. Chem. Phys., 16, 11497–11519, doi: 10.5194/acp-16-11497-2016, 2016.
- 5 Gilette, D.A.: Threshold Friction Velocities for Dust Production for Agricultural Soils. J. Geophys. Res. Atmos., 93, 12645-12662, doi: 10.1029/JD093iD10012645, 1988.
 - Govaerts, Y.M., Wagner, S., Lattanzio, A., and Watts P.: Joint retrieval of surface reflectance and aerosol optical depth from MSG/SEVIRI observations with an optimal estimation approach: 1. Theory, J. Geophys. Res.-Atmos., 115, D02203, doi:10.1029/2009JD011779, 2010.
- 10 Guerreiro, C.B.B., Foltescu, V., and de Leeuw, F.: Air quality status and trends in Europe, Atmos. Environ., 98, 376–384, doi: 10.1016/j.atmosenv.2014.09.017, 2014.
 - Guo, J.P., Zhai, P., Wu, L., Cribb, M., Li, Z., Ma, Z., Wang, F., Chu, D., Wang, P., and Zhang, J.: Precipitation and air pollution at mountain and plain stations in northern China: Insights gained from observations and modeling, J. Geophys. Res. -Atmos., 119 (8), 4793-4807, doi: 10.1002/2013jd021161 2014.
- 15 Guo, J., Deng, M., Lee, S. S., Wang, F., Li, Z., Zhai, P., Liu, H., Lv, W., Yao, W., and Li, X.: Delaying precipitation and lightning by air pollution over the Pearl River Delta. Part I: Observational analyses, J. Geophys. Res. Atmos., 121, 6472–6488, doi: 10.1002/2015JD023257, 2016a.
 - Guo, M., Sun, L., and Xu, X.: Inter-comparison of aerosol optical thickness from MODIS, MISR, and OMI using measurements from solar radiation stations in China, in: Proceedings of SPIE 9876, Remote Sensing of the Atmosphere, Clouds, and Precipitation VI, New Delhi, India, 6 September 2016, doi:10.1117/12.2228000, 2016b.

- Guo, J., Liu, H., Li, Z., Rosenfeld, D., Jiang, M., Xu, W., Jiang, J. H., He, J., Chen, D., Min, M., and Zhai, P.: Aerosol-induced changes in the vertical structure of precipitation: a perspective of TRMM precipitation radar, Atmos. Chem. Phys., 18, 13329-13343, https://doi.org/10.5194/acp-18-13329-2018, 2018.
- Hao, J., Wang, S., Liu, B., and He., K.: Designation of acid rain and SO2 control zones and control policies in China, J. Environ. Sci. Heal A, 35, 1901–1914, doi: 10.1080/10934520009377085, 2000.
- Hollmann, R., Merchant, C. J., Saunders, R., Downy, C., Buchwitz, M., Cazenave, A., Chuvieco, E., Defourny, P., de Leeuw, G., Forsberg, R., Holzer-Popp, T., Paul, F., Sandven, S., Sathyendranath, S., van Roozendael, M., and Wagner, W.: The ESA climate change initiative: satellite data records for essential climate variables, B. Am. Meteorol. Soc., 94, 1541-1552, doi: 10.1175/BAMS-D-11-00254.1, 2013.
- 30 Holzer-Popp, T., de Leeuw, G., Griesfeller, J., Martynenko, D., Klüser, L., Bevan, S., Davies, W., Ducos, F., Deuz é, J. L., Graigner, R. G., Heckel, A., von Hoyningen-Hüne, W., Kolmonen, P., Litvinov, P., North, P., Poulsen, C. A., Ramon, D., Siddans, R., Sogacheva, L., Tanre, D., Thomas, G. E., Vountas, M., Descloitres, J., Griesfeller, J., Kinne, S., Schulz, M., and Pinnock, S.: Aerosol retrieval experiments in the ESA Aerosol_cci project, Atmos. Meas. Tech., 6, 1919-1957, doi:10.5194/amt-6-1919-2013, 2013.

- Hsu, N. C., Tsay, S. -C., King, M. D., and Herman, J. R.: Aerosol properties over bright-reflecting source regions, IEEE T. Geosci. Remote, 42, 557-569, doi: 10.1109/TGRS.2004.824067, 2004.
- Hsu, N. C., Jeong, M.-J., Bettenhausen, C., Sayer, A. M., Hansell, R., Seftor, C. S., Huang, J., and Tsay, S.-C.: Enhanced Deep Blue aerosol retrieval algorithm: The second generation, J. Geophys. Res. Atmos., 118, 9296–9315, doi:10.1002/jgrd.50712, 2013.
- Hsu, N. C., Lee, J., Sayer, A. M., Carletta, N., Chen, S.-H., Tucker, C. J., Holben, B. N., and Tsay, S.-C.: Retrieving Near-Global Aerosol Loading over Land and Ocean from AVHRR, J. Geophys. Res. Atmos., 122, doi:10.1002/2017JD026932, 2017.
- Ichoku, C, D. A., Chu, D. A., Mattoo, S., Kaufman, Y. J., Remer, L. A., Tanre ´, D., Slutsker, I., and Holben, B. N.: A spatio-temporal approach for global validation and analysis of MODIS aerosol products, Geophys. Res. Lett., 29, MOD1-1-MOD1-4, doi: 10.1029/2001GL013206, 2002.

- Jin, Y., Andersson, H., and Zhang, S.: Air Pollution Control Policies in China: A Retrospective and Prospects, Int. J. Environ. Res. Public Health, 13, 1219, doi:10.3390/ijerph13121219, 2016.
- Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J. J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, Biogeosciences, 9, 527–554, https://doi.org/10.5194/bg-9-527-2012, 2012.
- Kang, H., Zhu, B., van der A. R., Zhu, C., de Leeuw, G., Hou, X., and Gao, J.: Natural and anthropogenic contributions to long-term variations of SO2, NO2, CO, and AOD over East China, Atmos. Environ., 215, 284-293, https://doi.org/10.1016/j.atmosres.2018.09.012, 2018.
- 20 Kaufman, Y. J., Wald, A. E., Remer, L. A., Gao, B. C., Li, R. R., and Flynn, L.: The MODIS 2.1-mu m channel Correlation with visible reflectance for use in remote sensing of aerosol, IEEE T. GEOSCI. REMOTE, 35, 1286-1298, doi: 10.1109/36.628795, 1997.
 - Koren, I., Dagan, G., and Altaratz, O.: From aerosol-limited to invigoration of warm convective clouds, Science, 344(6188), 1143-1146, 2014.
- 25 Kolmonen, P., Sogacheva, L., Virtanen, T.H., de Leeuw, G., and Kulmala, M.: The ADV/ASV AATSR aerosol retrieval algorithm: current status and presentation of a full-mission AOD data set, Int. J. Digit. Earth, 9:6, 545-561, doi: 10.1080/17538947.2015.1111450, 2016.
 - Levy, R. C., Remer, L. A., Kleidman, R. G., Mattoo, S., Ichoku, C., Kahn, R., and Eck, T. F.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmos. Chem. Phys. 10, 10399-10420, doi:10.5194/acp-10-10399-2010, 2010.
 - Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The Collection 6 MODIS aerosol products over land and ocean, Atmos. Meas. Tech., 6, 2989-3034, doi:10.5194/amt-6-2989-2013, 2013.

- Levy, R. C., Munchak, L. A., Mattoo, S., Patadia, F., Remer, L. A., and Holz, R. E.: Towards a long-term global aerosol optical depth record: applying a consistent aerosol retrieval algorithm to MODIS and VIIRS-observed reflectance, Atmos. Meas. Tech., 8, 4083-4110, https://doi.org/10.5194/amt-8-4083-2015, 2015.
- Li, J., Liu, R., Liu, S., Shiu, C., Wang, J., and Zhang, Y.: Trends in aerosol optical depth in northern China retrieved from sunshine duration data, Geophys. Res. Lett., 43, 431-439, doi: 10.1002/2015GL067111, 2016.
- Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu B.: Aerosol and boundary-layer interactions and impact on air quality, Natl. Sci. Rev., 4, 810–833, doi: 10.1093/nsr/nwx117, 2017a.
- Li, Z., Xu, H., Li, K. T., Li, D. H., Xie, Y. S., Li, L., Zhang, Y., Gu, X. F., Zhao, W., Tian, Q. J., Deng, R. R., Su, X. L., Huang, B., Qiao, Y. L., Cui, W. Y, Hu, Y., Gong, C. L., Wang, Y. Q., Wang, X. F., Wang, J. P., Du, W. B., Pan, Z. Q., Li, Z. Z., and Bu, D.: Comprehensive study of optical, physical, chemical and radiative properties of total columnar atmospheric aerosols over China: An overview of Sun-sky radiometer Observation NETwork (SONET) measurements, Bull. Amer. Meteor. Soc., doi:10.1175/BAMS-D-17-0133.1, 2018.

- Lu, S., Lin, H., Heemink, A., Segers, A., and Fu, G.: Estimation of volcanic ash emissions through assimilating satellite data and ground-based observations, J. Geophys. Res. Atmos., 121, 10971–10994, doi:10.1002/2016JD025131, 2016.
- Miao, Y., Guo, J., Liu, S., Liu, H., Zhang, G., Yan, Y., and He, J.: Relay transport of aerosols to Beijing-Tianjin-Hebei region by multi-scale atmospheric circulations, Atmos. Environ., 165, 35-45, doi: 10.1016/j.atmosenv.2017.06.032, 2017.
- Miller, D. J., Sun, K., Zondlo, M. A., Kanter, D., Dubovik, O., Welton, E. J., Winker, D. M., and Ginoux, P.: Assessing boreal forest fire smoke aerosol impacts on U.S. air quality: A case study using multiple data sets, J. Geophys. Res., 116, D22209, doi:10.1029/2011JD016170, 2011.
- O'Dowd, C.D., and de Leeuw, G.: Marine Aerosol Production: a review of the current knowledge, Phil. Trans. R. Soc. A, 365, 1753–1774, doi:10.1098/rsta.2007.2043, 2007.
- Oertel, C., Matschullat, J., Zurba, K., Zimmermann, F., and Erasmi, S.: Greenhouse gas emissions from soils A review, Chem. Erde Geochem., 76, 327–352. http://dx.doi.org/10.1016/j.chemer.2016.04.002, 2016.
- 25 Popp, T., de Leeuw, G., Bingen, C., Brühl, C., Capelle, V., Chedin, A., Clarisse, L., Dubovik, O., Grainger, R., Griesfeller, J., Heckel, A., Kinne, S., Klüser, L., Kosmale, M., Kolmonen, P., Lelli, L., Litvinov, P., Mei, L., North, P., Pinnock, S., Povey, A., Robert, C., Schulz, M., Sogacheva, L., Stebel, K., Stein Zweers, D., Thomas, G., Tilstra, L.G., Vandenbussche, S., Veefkind, P., Vountas, M., and Xue, Y.: Development, production and evaluation of aerosol Climate Data Records from European satellite observations (Aerosol_cci), Remote Sens., 8, 421, doi:10.3390/rs8050421, 2016.
- 30 Qiu, J.: A Method to Determine Atmospheric Aerosol Optical Depth Using Total Direct Solar Radiation, J. Atmos. Sci., 55, 744-757, doi: 10.1175/1520-0469(1998)055<0744:AMTDAA>2.0.CO;2, 1998.
 - Remer, L. A., Kaufman, Y. J., Tanr & D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R.-R., Ichoku, C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B. N.: The MODIS aerosol algorithm, products, and validation, J. Atmos. Sci., 62, 947–973, doi: http://dx.doi.org/10.1175/JAS3385.1, 2005.

- Riffler, M., Popp, C., Hauser, A., Fontana, F., and Wunderle, S.: Validation of a modified AVHRR aerosol optical depth retrieval algorithm over Central Europe, Atmos. Meas. Tech., 3, 1255-1270, 2010.
- Robel, J., and Graumann, A.: NOAA KLM USER'S GUIDE with NOAA-N, N Prime, and MetOp SUPPLEMEN: https://www.nasa.gov/pdf/111742main_noaa_n_booklet.pdf, 2014.
- 5 Salomonson, V. V., Barnes, W. L., Maymon, P. W., Montgomery, H. E., Ostrow, H.: MODIS Advanced facility instrument for studies of the earth as a system, IEEE T. Geosci. Remote, 27, 145-153, doi: 10.1109/36.20292, 1989.
 - Sayer, A. M., Hsu, N. C., Bettenhausen, C., and Jeong, M.-J.: Validation and uncertainty estimates for MODIS Collection 6 "Deep Blue" aerosol data, J. Geophys. Res. Atmos., 118, 7864–7872, doi:10.1002/jgrd.50600, 2013.
 - Sayer, A. M., Munchak, L. A., Hsu, N. C., Levy, R. C., Bettenhausen, C., and Jeong, M.-J.: MODIS Collection 6 aerosol products: Comparison between Aqua's e-Deep Blue, Dark Target, and "merged" data sets, and usage recommendations, J. Geophys. Res. Atmos., 119, 13965–13989, doi: 10.1002/2014JD022453, 2014.

25

- Sayer, A. M., Hsu, N. C., Bettenhausen, C., Jeong, M.-J., and Meister, G.: Effect of MODIS Terra radiometric calibration improvements on Collection 6 Deep Blue aerosol products: Validation and Terra/Aqua consistency, J. Geophys. Res. Atmos., 120, 12157-12174, doi: 10.1002/2015JD023878, 2015.
- Sayer, A. M., Hsu, N. C., Lee, J., Carletta, N., Chen, S. -H., and Smirnov, A.: Evaluation of NASA Deep Blue/SOAR aerosol retrieval algorithms applied to AVHRR measurements, J. Geophys. Res. Atmos., doi: 10.1002/2017JD026934, doi:10.1002/2017JD026934, 2017.
 - Seinfeld, J. H., Pandis, S. N., and Noone, K.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Phys. Today, 51, 88, doi: 10.1063/1.882420, 1998.
- 20 Sogacheva, L., Kolmonen, P., Virtanen, T. H., Rodriguez, E., Saponaro, G., and de Leeuw, G.: Post-processing to remove residual clouds from aerosol optical depth retrieved using the Advanced Along Track Scanning Radiometer, Atmos. Meas. Tech., 10, 491-505, doi:10.5194/amt-10-491-2017, 2017.
 - Sogacheva, L., de Leeuw, G., Rodriguez, E., Kolmonen, P., Georgoulias, A. K., Alexandri, G., Kourtidis, K., Proestakis, E., Marinou, E., Amiridis, V., Xue, Y., and van der A, R. J.: Spatial and seasonal variations of aerosols over China from two decades of multi-satellite observations Part 1: ATSR (1995–2011) and MODIS C6.1 (2000–2017), Atmos. Chem. Phys., 18, 11389-11407, https://doi.org/10.5194/acp-18-11389-2018, 2018a.
 - Sogacheva, L., Rodriguez, E., Kolmonen, P., Virtanen, T. H., Saponaro, G., de Leeuw, G., Georgoulias, A. K., Alexandri, G., Kourtidis, K., and van der A, R. J.: Spatial and seasonal variations of aerosols over China from two decades of multisatellite observations. Part II: AOD time series for 1995–2017 combined from ATSR ADV and MODIS C6.1 for AOD tendencies estimation, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-288, in review, 2018b.
 - Stark, M. S., Harrison, J. T. H., Anastasi, C.: The formation of nitrogen oxides by electrical discharges and implications for atmospheric lightning, J. Geophys. Res.-Atmos., 101, 6963-6969, doi: 10.1029/95JD03008, 1996.

- Stavrakou, T., Müller, J. F., Bauwens, M., De Smedt, I., Lerot, C., Van Roozendael, M., Coheur, P. F., Clerbaux, C., Boersma, K. F., van der A. R., and Song Y.: Substantial Underestimation of Post Harvest Burning Emissions in the North China Plain Revealed by Multi Species Space Observations, Sci. Rep. UK, 6, 32307, doi: 10.1038/srep32307, 2016.
- Stowe, L. L., McClain, E. P., Carey, R., Pellegrino, P., Gutman, G. G., Davis, P., Long, C., and Hart, S.: Global distribution of cloud cover derived from NOAA/AVHRR operational satellite data, Adv. Space Res., 11, 51–54, doi: 10.1016/0273-1177(91)90402-6. 1991.
 - Stowe, L.L., Davis, P.A., and McClain, E.P.: Scientific basis and initial evaluation of the CLAVR-1 global clear cloud classification algorithm for the advanced very high resolution radiometer, J. Atmos. Ocean Technol., 16, 656–681, https://doi.org/10.1175/1520-0426(1999)016<0656:SBAIEO>2.0.CO;2, 1999.
- 10 Sundström, A.-M., Kolmonen, P., Sogacheva L., and de Leeuw, G.: Aerosol retrievals over China with the AATSR Dual-View Algorithm, Remote Sens. Environ., 116, 189-198, doi: 10.1016/j.rse.2011.04.041, 2012.
 - Tanré, D., Kaufman, Y. J., Herman, M., and Mattoo, S.: Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances, J. Geophys. Res. Atmos., 102, 16971-16988, doi: 10.1029/96JD03437, 1997.
- Theys, N., Campion, R., Clarisse, L., Brenot, H., van Gent, J., Dils, B., Corradini, S., Merucci, L., Coheur, P.-F., Van Roozendael, M., Hurtmans, D., Clerbaux, C., Tait, S., and Ferrucci, F.: Volcanic SO2 fluxes derived from satellite data: a survey using OMI, GOME-2, IASI and MODIS, Atmos. Chem. Phys., 13, 5945-5968, https://doi.org/10.5194/acp-13-5945-2013, 2013.
 - van der A, R. J., Mijling, B., Ding, J., Koukouli, M. E., Liu, F., Li, Q., Mao, H., and Theys, N.: Cleaning up the air: effectiveness of air quality policy for SO2 and NOx emissions in China, Atmos. Chem. Phys., 17, 1775-1789, doi:10.5194/acp-17-1775-2017, 2017.

- Veefkind, J.P., de Leeuw, G., and Durkee, P.A: Retrieval of aerosol optical depth over land using two-angle view satellite radiometry during TARFOX, Geophys. Res. Letters, 25, 3135-3138, doi: 10.1029/98GL02264, 1998.
- WHO (2018), World Health Organization: http://www.who.int/phe/health_topics/outdoorair/databases/en/, last accessed 16 July 2018.
- Wang, Y., Yao, L., Wang, L., Liu Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Sci. China Earth Sci., 57, 12-25, doi: 10.1007/s11430-013-4773-4, 2013.
 - Xin, J., Wang, Y., Pan. Y., Ji, D., Liu, Z., Wen, T., Wang, Y., Li, X., Sun, Y., Sun J., Wang P., Wang G., Wang X., Cong Z., Song T., Hu B., Wang L., Tang G., Gao W., Guo Y., Miao H., Tian S., and Wang L., The Campaign on Atmospheric Aerosol Research Network of China: CARE-China, Bull. Amer. Meteor. Soc., 96, 1137–1155, doi: 10.1175/BAMS-D-14-00039.1. 2015.
 - Xu, X., Qiu, J., Xia, X., Sun, L., and Min M.: Characteristics of atmospheric aerosol optical depth variation in China during 1993-2012, Atmos. Environ., 119, 82-94, doi: 10.1016/j.atmosenv.2015.08.042, 2015.

- Xue, Y., He, X., de Leeuw, G., Mei, L., Che, Y., Rippin, W., Guang, J., and Hu, Y.: Long-time series aerosol optical depth retrieval from AVHRR data over land in North China and Central Europe, Remote Sens. Environ., 198, 471-489, doi: 10.1016/j.rse.2017.06.036, 2017.
- Yim S.H.L., Hou X., Guo J., Yang Y.: Contribution of local emissions and transboundary air pollution to air quality in Hong Kong during El Niño Southern Oscillation and heatwaves, Atmos. Res., 218, 50-58, https://doi.org/10.1016/j.atmosres.2018.10.021, 2019.

- Yu, Y., Kalashnikova, O. V., Garay, M. J., Lee, H., and Notaro, M.: Identification and characterization of dust source regions across North Africa and the Middle East using MISR satellite observations, Geophys. Res. Lett., 45, doi: 10.1029/2018GL078324, 2018.
- 10 Zhang, L., Liu, Y. and Hao, L.: Contributions of open crop straw burning emissions to PM2.5 concentrations in China. Environ. Res. Lett., 11, 014014, doi:10.1088/1748-9326/11/1/014014, 2016.
 - Zhang, J., Reid, J.S., Alfaro-Contreras, R., and Xian, P.: Has China been exporting less particulate air pollution over the past decade? Geophys. Res. Lett., 44, 2941–2948, doi:10.1002/2017GL072617, 2017.
- Zhao, T. X.-P., Laszlo, I., Guo, W., Heidinger, A., Cao, C., Jelenak, A., Tarpley, D., and Sullivan, J.: Study of long-term trend
 in aerosol optical thickness observed from operational AVHRR satellite instrument, J. Geophys. Res., 113, D07201, doi:10.1029/2007JD009061, 2008.
 - Zhao, B., Jiang, J.H., Gu, Y., Diner, D., Worden, J., Liou, K.-N., Su, H., Xing, J., Garay, M., and Huang, L.: Decadal-scale trends in regional aerosol particle properties and their linkage to emission changes, Environ. Res. Lett., 12, 054021(2017), doi.org/10.1088/1748-9326/aa6cb2, 2017.
- 20 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-374, in review, 2018.

Appendix A

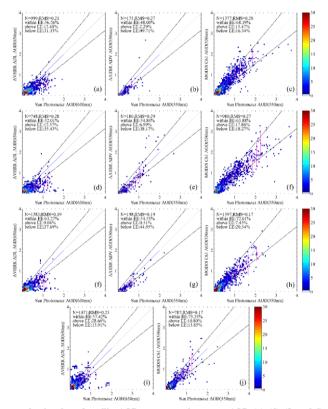


Figure A1: Seasonal scatter density plots of satellite AOD versus sun photometer AOD. (a), (d), (f), and (i) are AVHRR AOD vs. AERONET AOD in Spring, Summer, Autumn, and Winter; (b), (e), and (g) are ATSR AOD vs. AERONET AOD in Spring, Summer, and Autumn; (c), (f), (h), and (j) are MODIS AOD vs. AERONET AOD in Spring, Summer, Autumn, and Winter.

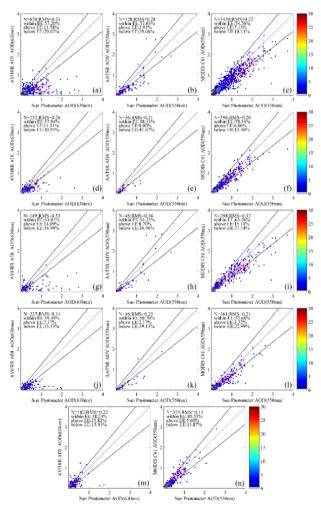


Figure A2: Scatter density plots of satellite-retrieved AOD versus <u>sunphotometersun photometer</u> retrieved AOD data from Beijing station. ADL all data (a), in Spring (d), in Summer (g), in Autumn (j), and in Winter (m) vs. AERONET, ADV all data (b), in Spring (e), in Summer (h), and in Autumn (k) vs. AERONET, MODIS all data (c), in Spring (f), in Summer (i), in Autumn (l), and in Winter (n) vs. AERONET.

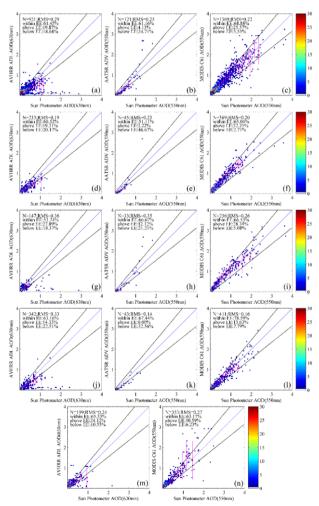


Figure A3: Scatter density plots of satellite-retrieved AOD versus <u>sumphotometer sum photometer</u> retrieved AOD data from XiangHe station. ADL all data (a), in Spring (d), in Summer (g), in Autumn (j), and in Winter (m) vs. AERONET, ADV all data (b), in Spring (e), in Summer (h), and in Autumn (k) vs. AERONET, MODIS all data (c), in Spring (f), in Summer (i), in Autumn (l), and in Winter 5 (n) vs. AERONET.

Investigations into the Development of a Satellite-Based Aerosol Climate Data Record using ATSR-2, AATSR and AVHRR data over North-Eastern China from 1987 to 2012

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Abstract. Satellites provide information on the temporal and spatial distributions of aerosols on regional and global scales. With the same method applied to a single sensor all over the world, a consistent data set is to be expected. However, the application of different retrieval algorithms to the same sensor, and the use of a series of different sensors may lead to substantial differences and no single sensor or algorithm is better than any other everywhere and at any time. For the production of long-term climate data records, the use of multiple sensors cannot be avoided. The Along Track Scanning Radiometer (ATSR-2) and the advanced ATSR (AATSR) Aerosol Optical Depth (AOD) data sets have been used to provide a global AOD data record over land and ocean of 17-years (1995-2012), which is planned to be extended with AOD retrieved from a similar sensor. To investigate the possibility to extend the ATSR data record to earlier years, the use of an AOD data set from the Advanced Very High Resolution Radiometer (AVHRR) is investigated. AOD data sets used in this study were retrieved from the ATSR sensors using the ATSR Dual View algorithm ADV v2.31 developed by Finnish Meteorological Institute (FMI), and from the AVHRR sensors using the ADL algorithm developed by RADI/CAS. Together these data sets cover a multidecadal period (1987-2014). The study area includes two contrasting areas, both as regards aerosol content and composition and surface properties, i.e. a region over North-East (NE) China encompassing a highly populated urban/industrialized area (Beijing-Tianjin-Hebei) and a sparsely populated mountainous area.

Ground-based AOD observations available from ground-based sun photometer AOD data in AERONET and CARSNET are used as reference, together with broadband extinction methods (BEM) data at Beijing to cover the time before sun photometer observations became available in the early 2000's. In addition, MODIS-Terra C6.1 AOD data are used as reference data set over the wide area where no ground-based data are available. All satellite data over the study area were validated versus the reference data, showing the qualification of MODIS for comparison with ATSR and AVHRR. The comparison with MODIS shows that AVHRR performs better that ATSR in the north of the study area (40 N), whereas further south ATSR provides

better results. The validation versus sun photometer AOD shows that both AVHRR and ATSR underestimate the AOD, with ATSR failing to provide reliable results in the winter time. This is likely due to the highly reflecting surface in the dry season, when AVHRR-retrieved AOD traces both MODIS and reference AOD data well. However, AVHRR does not provide AOD larger than about 0.6 and hence is not reliable when high AOD values have been observed over the last decade. In these cases, ATSR performs much better, for AOD up to about 1.3. AVHRR-retrieved AOD compares favourably with radiance-derived AOD, except for AOD higher than about 0.6. These comparisons lead to the conclusion that AVHRR and ATSR AOD data records each have their strengths and weaknesses which need to be accounted for when combining them in a single multi-decadal climate data record.

1 Introduction

30

Aerosol particles are important atmospheric constituents, which play significant roles in many processes such as atmospheric chemistry, the absorption and scattering of solar radiation, and the lifetime of cloud and precipitation systems (Boucher et al, 2013; Koren et al., 2014; Guo et al., 2014; 2016a; 2018). Aerosol particles have an adverse effect on human health and are responsible for 7 million premature deaths annually over the whole world (WHO, 2018). Processes involving aerosols and their effects depend on the chemical and physical properties of the aerosol particles which in turn are determined by sources of directly emitted particles, the formation of aerosols from precursor gases (and thus the sources of these gases), the transformation of these particles during chemical and physical processes in the atmosphere and their removal by wet or dry deposition (cf. Seinfeld and Pandis, 1998, for a comprehensive treatment of aerosol processes). Observations of the concentrations of trace gases and aerosols are publicly available since several observational networks have been established, such as NASA's AERONET (AErosol RObotic NETwork) (Holben et al. 1998) (with observations mainly in the east of China), CARE-China (Xin et al., 2015), the Chinese Aerosol Remote Sensing Network (CARSNET) (Che et al., 2009; 2015) and SONET (Sun-sky radiometer Observation NETwork) (Li et al., 2018). However, most of these observations started in the last decade and very few, if any, historical data on large scale are available for the construction of the long time series needed to show the evolution of pollutant concentrations over many years and analyse the effects of different contributions. Here, satellite data may offer a solution. The most common satellites used for the observation of trace gases and aerosols offer information since the beginning of the 21st century and, by combining the information from different instruments, time series encompassing two decades can be constructed (de Leeuw et al., 2018, Sogacheva et al., 2018b). Satellite information has been used together with model simulation to analyse the effects of natural and anthropogenic contributions on the concentrations of trace gases and aerosols (Kang et al., 2018). In another study combining satellite data with ground-based observations, the role of precursor gases, in particular VOCs, and photochemical reactions in the formation of aerosols (PM2.5) was revealed (Bai et al., 2018).

In the second half of the 20th century the adverse effects of the precursor gases like SO₂ and NO₂ and aerosols on climate, air quality, and health were recognized and reduced by effective measures in developed countries. These led to the reduction of

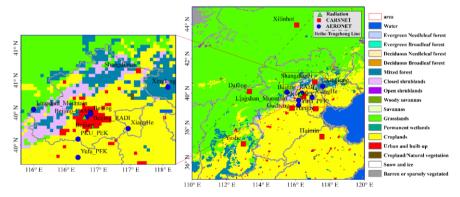
air pollution in developed countries, in particular in North America and Europe (Guerreiro et al., 2014), but in developing countries with increasing industrial activity and urbanization the concentrations continued to increase (Hao et al., 2000). As an example, in China the concentrations of pollutants have increased over the years and are amongst the highest in the world. Recent publications show the effect of policy measures on the reduction of the most polluting trace gases SO₂ and NO₂ (van der A et al., 2017), which, as precursor gases, also affect the concentrations of aerosols. In particular, the emissions of SO₂ were reduced as part of the 11th Five-Year Plan (2006-2010) (Zheng et al., 2018), but the emissions of NO₂ continued to increase (e.g., van der A et al., 2017) until the 12th Five-Year Plan (2011-2015). Large emission reductions were achieved after 2013 when the Clean Air Action was enacted and implemented and the NO₂ concentrations decreased (Zheng et al., 2018). Starting from 2011, aerosol concentrations decreased in China as shown, e.g., from satellite observations of the aerosol optical depth (AOD) (Zhang et al., 2017, Zhao et al., 2017, de Leeuw et al, 2018, Sogacheva et al., 2018b). However, early policies before 2005 on reducing pollution emissions are not effective, as shown from sparse ground-based observations (Jin et al., 2016). Long term satellite retrievals may offer a solution on a large spatial scale to observe how air pollutions control policies work and air pollution changes.

The Along Track Scanning Radiometer (ATSR-2), a dual view instrument, was launched in 1995 on the European Space Agency (ESA) satellite ERS-2 and provided data until 2003, and was one of the earliest satellite instruments used to retrieve Aerosol Optical Depth (AOD) quantitatively (Flowerdew and haigh, 1996; Veefkind et al., 1998). Its successor, the Advanced ATSR (AATSR) is a similar instrument launched in 2002 on the ESA platform ENVISAT, which was lost in April 2012. The AOD over China from ATSR-2 and AATSR is consistent (Sogacheva et al., 2018a) and hence, together these instruments provide a 17-year AOD time series, 1995-2012 (Popp et al., 2016, de Leeuw et al., 2018). Combining ATSR and MODIS, 22year AOD measurements were constructed, showing how the AOD increased until about 2006, and then clearly decreased since 2011 over China (de Leeuw et al., 2018, Sogacheva et al., 2018b). However, this time series should be extended especially before the 1990s when great changes have taken place in aerosols in China with the increase of industrialization.. The Advanced Very High Resolution Radiometer (AVHRR) onboard the NOAA satellites series could be a good choice to extend the ATSR time series as it started observations continuously from 1978 to the present. Xue et al. (2017) developed an AOD data set encompassinges two relatively small areas over Europe and China from 1983 to 2014, but covers the complete period, as opposed to the AVHRR global over land AOD data set recently presented by Hsu et al. (2017) and Sayer et al. (2017) which encompasses several distinct time periods. Hence in this study we focus here on the Xue et al (2017) AVHRR AOD data set over China and compare that with ATSR-derived AOD data to determine its suitability for merging (as done for ATSR/MODIS by Sogacheva et al. 2018b), and thus extending the ATSR data set before the ATSR-2 era (and possibly after AATSR was lost in 2012, although other data sets may be more suitable for to extend to later years as shown in Sogacheva et al. (2018b) for MODIS. So the focus of the current study is to investigate whether the ATSR AOD data set can be extended to earlier years by using the Xue et al. (2017) AVHRR AOD data over China. For comparison of the ATSR and AVHRR AOD data sets, we use both ground-based reference data, from AERONET (Holben et al., 1988) and from CARSNET (Che et al, 2009, 2015), and MODIS C6.1 AOD data. These reference data are not available for the period before 2000 and therefore we also use AOD data derived from broadband radiation measurements using the broadband extinction method (BEM) (Xu et al., 2015, Guo et al, 2016b) as described in Sect. 2.3.3. Data sets and methods used are presented in section 2. An overview of the data and an evaluation of their quality are presented in section 3, including a comparison of the various data sets. The results are discussed and conclusions are presented in section 4.

2 Method

2.1 Study area

The study area is located over north-eastern China, i.e. between 110 ° and 120 °E and 35 ° and 45 °N (Fig. 1), which is divided into two sub-regions by the Taihang Mountain range with the North China Plain (NCP) and large urban agglomerations like Beijing and Tianjin and the Hebei province (together BTH which is among the most populated and fast developing regions in China) to the SE and the mountainous terrain to the NW extending over the Loess Plateau in Shanxi Province and the Inner Mongolia plateau. The Taihang mountain range forms a natural barrier for the transport of air pollution resulting in the frequent accumulation of pollutants and the occurrence of haze over the BTH area and the NCP (e.g., Sundström et al., 2012; Wang et al., 2013). The satellite-derived AOD maps in Fig. 2 show that this line also roughly divides high AOD in the SW of the study area and low AOD in the NW. The background in Fig. 1 is a land cover map showing that the major land cover types in the study area are croplands in the SE and grassland to the NW, which are intersected by mixed forest and closed shrublands as shown in the inset in Fig. 1.



6 Figure 2: Study area, with the locations of the ground-based reference sites discussed in Sect. 2.3 (CARSNET: red squares, AERONET: blue circles, solar radiation station: green triangle) overlaid on the IGBP land cover map.

2.2 Satellite data

Satellite-retrieved data sets used in this study are satellite-derived AOD data from six radiometers, i.e. AVHRR-1, AVHRR-2, AVHRR-3, ATSR-2, AATSR and MODIS. These data sets are briefly discussed below.

2.2.1 AVHRR

The AVHRR instruments flew on a series of satellites, most of them with an afternoon equator crossing time at 13:40 LT (ascending) (see Xue et al., 2017, for an overview). AVHRR has a swath width of 2399 km at 833km altitude (Robel and Graumann, 2014) and thus provides daily global coverage. The AVHRR sensor was designed for measuring cloud cover and surface temperature, but the observations are also used for the retrieval of AOD over ocean (e.g. Zhao et al., 2008), with only few efforts to retrieve the AOD over land. Recently two AOD data sets retrieved from AVHRR observations over land were published, the first one by Xue et al. (2017) and short after another one by Sayer et al. (2017) and Hsu et al (2017). Xue et al. (2017) used the Algorithm for the retrieval of the aerosol optical Depth over Land (ADL) which was used to produce the continuous data set over China used in the current study. The use of AVHRR data for aerosol retrieval requires a re-calibration of the radiances measured at the top of the atmosphere (TOA) because the sensors have no in-flight calibration. Xue et al. (2017) describe how this was achieved. Major problems in the retrieval of AOD from satellite observations is the effective decoupling of atmospheric and surface effects on the reflectance measured at the top of the atmosphere (TOA), cloud detection and the description of the aerosol properties. In the ADL algorithm a cloud mask is applied based on the Clouds from AVHRR (CLAVR, Stowe et al., 1991, 1999) and in the retrieval six aerosol types are used as proposed by Govaerts (2010). For cloudfree pixels, and after application of gas absorption corrections as described in Xue et al. (2017), the land surface reflectance in the AVHRR channel 1 (0.64 µm) is parameterized in terms of the measured reflectance in Channel 3 (3.75 µm), with coefficients which are functions of the NDVI and scattering angle. The aerosol extinction at 3.75 µm is assumed to be negligible (which may not be true in the presence of coarse aerosol particles such as for desert dust). It is noted that the TOA reflectance at 3.75 µm is composed of solar radiation reflected by the land/atmosphere system and Earth radiation. The reflected part is estimated by using the method proposed by Allen et al. (1990). For each land cover type, as determined following the International Geosphere-Biosphere Programme (IGBP) land cover classification, a different parameterization of the 0.64 µm reflectance was developed. The IGBP land cover information used is the MODIS MCD12C1 product obtained from https://lpdaac.usgs.gov/dataset_discovery/modis/modis_products_table/mcd12c1, and is described in the MOD12 product ATBD (https://modis.gsfc.nasa.gov/data/atbd/atbd_mod12.pdf).

In the implementation of the ADL algorithm, the land surface reflectance is coupled to a radiative transfer model which includes individual parameterizations of the direct, single-scattered and multiple-scattered radiances. Thus a model is developed for the TOA reflectance which is solved by optimal estimation. For more detail, see Xue et al., (2017). AOD is retrieved at a wavelength of 0.64 μ m at a spatial resolution of 0.05 °× 0.05 °. In this study AOD retrieved from NOAA-10 (1987-1991), NOAA-12 (1992-1998), NOAA-15 (1999-2002), NOAA-17 (2003-2009), and METOP-A (2010-2014) is used.

The AVHRR data from 1983 to 1986 are not used because there are Jan to May in 1983, May to Jul in 1984, Jul to Oct in 1985, and Nov to Dec in 1986 are available, not consistent.

2.2.2 ATSR (ATSR-2 and AATSR)

Two Along-Track Scanning Radiometers (together referred to as ATSR) are used in this study: the ATSR-2, which flew on ESA's ERS-2 from 1995-2003, and the Advanced ATSR (AATSR), which flew on ESA's environmental satellite ENVISAT and provided data from May 2002 until April 2012. Both satellites flew in a sun-synchronous descending orbit with a day-time equator crossing time of 10:30 LT (ERS-2) and 10:00 LT (ENVISAT). Together these instruments provided 17 years of global aerosol data. The ATSR sensor has two views (near-nadir and 55° forward from nadir) which facilitate effective separation of the surface and atmospheric contributions to the reflectance at TOA. Multiple wavelengths (7) from VIS to TIR facilitate effective cloud screening and allow for multi-wavelength retrieval of aerosol properties. ATSR has a conical scan mechanism with a swath of 512 km, resulting in daily global coverage in 5-6 days.

The ATSR dual view was first applied for AOD retrieval over land by Veefkind et al. (1998), based on the principles outlined by Flowerdew and Haigh (1995). Over ocean the two views are used separately to retrieve the AOD in both the nadir and forward directions. The ATSR dual view algorithm (ADV) has been much improved during algorithm experiments (Holzer-Popp et al., 2013) as part of the ESA Climate Change Initiative (cci) (Hollmann et al., 2013) project Aerosol_cci (de Leeuw et al., 2015, Popp et al., 2016). The most recent version of the ATSR dual view algorithm ADV is described in Kolmonen et al. (2016). ATSR L1 gridded brightness temperature data are provided with a nominal resolution of 1x1 km² sub-nadir and aerosol data are provided at a default spatial resolution of 10×10 km² on a sinusoidal grid (L2) and at 1°x 1° (L3).

The ATSR product used in this paper is the AOD at a wavelength of 550 nm, over the study area for the full ATSR mission. The data were produced using ADV version 2.31 which includes cloud post-processing as described in Sogacheva et al. (2017). ATSR-2 retrieved AOD data are available for the period June 1995- December 2003, with some gaps in 1995 and 1996. AATSR data are available for the period May 2002 - April 2012, but some data are missing in 2002 (see de Leeuw et al., 2018, for more details).

2.2.3 MODIS

- 25 The MODerate resolution Imaging Spectroradiometer (MODIS) flies aboard the NASA Terra and Aqua satellites, launched in December 1999 and May 2002, respectively, in a near-polar sun-synchronous circular orbit with an equator crossing time of 10:30 and 13:30 (LT), respectively (Salomonson et al., 1989). MODIS is a single view instrument with a swath of 2330 km (cross track) and provides near-global coverage on a daily basis. One of the most successful products of MODIS, which has been used in numerous aerosol related studies, is the AOD at 550 nm.
- MODIS AOD is retrieved using two separate algorithms, Dark Target (DT) and Deep Blue (DB). In fact, two different DT algorithms are utilized, one for retrieval over land (vegetated and dark-soiled) surfaces (Kaufman et al., 1997, Remer et al.,

2005, Levy et al., 2010, 2013) and one for retrieval over water surfaces (Tanr é et al., 1997, Remer et al., 2005, Levy et al., 2013). The DB algorithm (Hsu et al., 2004, 2013) was traditionally used over bright surfaces where DT cannot be used (e.g. deserts, arid and semi-arid areas). However, the enhanced DB algorithm is capable of returning aerosol measurements over all land types (Sayer et al., 2013, 2014). The DT Expected Error (EE) is $\pm (0.05 + 0.15 \tau_{AERONET})$ over land and $\pm (0.04 + 0.1 \tau_{AERONET})$, $\pm (0.02 + 0.1 \tau_{AERONET})$ over sea relative to the AERONET optical thickness ($\tau_{AERONET}$) (Levy et al., 2013). The DB expected error is $\pm (0.03 + 0.2 \tau_{MODIS})$ relative to the MODIS optical thickness (τ_{MODIS}) (Hsu et al., 2013, Sayer et al., 2015). In this study the recently released (end of 2017) MODIS/Terra merged C6.1 L2 ($10x10 \text{ km}^2$) AOD dataset is used because of the proximity of the MODIS/Terra and AVHRR overpasses. The merged (DT and DB) dataset is described by Levy et al. (2013) and includes measurements from both algorithms, offers a better spatial coverage and can be used in quantitative scientific applications (Sayer et al., 2014). This merged data set was validated over China using AERONET data, showing the good performance for all retrieved AOD values up to 2.4, with a small overall bias of 0.06 (Sogacheva et al., 2018a).

2.3 Ground-based reference data

Ground-based reference data used in this study are sun photometers from AERONET (Holben et al., 1998) and CARSNET (Che et al., 2015) available in the study area (see Fig. 1), complemented with Broadband Extinction Method (BEM) AOD data in Beijing for the period when no sun photometer data are available. The eight AERONET sites in the study area are all located in, or close to, Beijing. Three of them (Beijing, Beijing_RADI, and PKU_PEK) are located in the city, the others are located in rural areas in the vicinity of Beijing. The eight CARSNET sites are distributed over a wider area extending beyond BTH, with two of them (Xilinhot and Datong) located to the NW of the Heihe-Tengchong line, Huimin in the NCP and Yushe in rural areas surrounded by mountains. The other four (Lingshan_Mountain, XiangHe, Xinglong, and Yufa_PEK) are in and around Beijing. The solar radiation station is located in Beijing, close to three AERONET sites (Beijing, Beijing_RADI. Beijing_CMA).

The AERONET, CARSNET and radiation-derived AOD data sets are briefly described below.

2.3.1 AERONET

The AERONET project is a federation of ground-based remote sensing aerosol networks established by the National Aeronautics and Space Administration (NASA) in the USA and PHOtométrie pour le Traitement Opérationnel de Normalisation Satellitaire (PHOTONS) in France. AERONET was expanded with other networks and national efforts, see the AERONET website (https://aeronet.gsfc.nasa.gov/) for a description of contributors, sites, operational procedures, data products and availability, etc. AERONET serves as the primary network for global validation of satellite-retrieved aerosol products, including AOD which is used in this study. AOD at all AERONET stations is measured using CIMEL sun photometers. Most common are the CIMEL CE-318 models with 5 wavelengths (440, 670, 870, 936 and 1020 nm) or polarized with 8 wavelengths, measuring direct sun as well at many angles using different scan patterns which provide the data necessary

to retrieve a multitude of aerosol properties. Data checking and processing is done centrally and the products are freely available from the AERONET website. Data are made available at three levels, i.e. Level 1.0 (unscreened), Level 1.5 (cloud-screened), and Level 2.0 (L2) (cloud-screened and quality-assured). The uncertainty of the CIMEL-derived AOD is 0.01-0.02 (wavelength dependent) (Eck et al., 1999). In this study we use AERONET L2 data from the most recent V3 dataset for validation and comparison (Giles et al., 2018). Satellite AOD data are commonly made available at 550 nm, and hence, for validation, the AERONET data are interpolated to this wavelength by using the Ångström Exponent (AE) which describes the AOD wavelength dependence (Ångström, 1924).

Almost all AERONET sites in the study area started observations after 2000 as Tab. 1 shows. Long-time and continuous measurements from 2001 to 2014 are only available from the Beijing and XiangHe stations. Therefore, we selected these stations for comparison with satellite time series, but for validation all available data are used.

Table 1. Data availability from AERONET sites in the study area (35 ° 45 °N, 110 ° 120 °E)

-	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
Beijing	√	√	√	√	√	√	√	√	√	√	√	√	√	√
Beijing-CAMS												√	√	√
Beijing_RADI										√	\checkmark			
Lingshan_Mountain														√
XiangHe	√			\checkmark	√	√	\checkmark	√	\checkmark	√	√	√	√	√
Xinglong						√	√	√	√	√	\checkmark	√		√
PKU_PEK						√		√						
Yufa_PEK						√								

2.3.2 CARSNET

AERONET sites in the study area are all located in or around Beijing (cf. Fig. 1). To expand the reference data set to other regions, AOD data from CARSNET (Che et al., 2015) was used, but only data for the years 2007, 2008 and 2010 were available for this study. The locations of the CARSNET sites in the study area are indicated in Fig. 1. CARSNET was established by the China Meteorological Administration (CMA) for the study of aerosol optical properties and validation of satellite retrievals, and the same instrumentation as AERONET (i.e. CIMEL CE-318) and similar procedures (Che et al., 2015). The difference is the way of instrument calibration and AOD calculation as described in Che et al. (2009). The CARSNET AOD uncertainties are 0.03, 0.01, 0.01 and 0.01 at the 1020, 870, 670 and 440 nm channels (Che et al., 2009).

2.3.3 Broadband extinction method (BEM) AOD data

Broadband solar radiation has been measured in China by China Meteorological Administration (CMA) since the 1950s at 98 sites (Qiu, 1998) which evolved into the China national solar radiation network with 14 stations (Xu et al., 2015) providing continuous data since 1993 using China-made pyrheliometers. Methods were developed to retrieve monochromatic or equivalent AOD using hourly accumulated direct solar radiation and the results of these BEM AOD are in good agreement with sun photometer data (see Xu et al., 2015 for an overview). In the current study we used BEM AOD at 550 nm retrieved from the pyrheliometer measurements at the Beijing station (Fig. 1) of the China national solar radiation network. These data are particularly useful for evaluation of the AVHRR-retrieved AOD for the period before 2000 when no sun photometer data were available. The AOD was retrieved using BEM described by Xu et al. (2015). Xu et al. (2015) evaluate the quality of the BEM AOD (at 750 nm) for the Beijing station from comparison with AERONET AOD. The results show that 59% of the hourly AOD data fall within an error envelope of $\pm (0.05 \pm 0.15\tau_{AERONET})$. For monthly averaged data this is 82%. Guo et al (2016) used the hourly mean BEM AOD at 550 nm to evaluate the products from MODIS, OMI and MISR, obtaining consistent results with previous validations based on sun photometer measurements, which proved the effectiveness of the BEM AOD in satellite product validation.

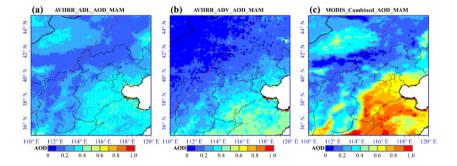
15 3 Result

3.1 Data overview

The aim of the present study is to determine the compatibility of AVHRR and ATSR AOD data sets and their suitability to combine them to extend the ATSR time series to the 1980s. AVHRR AOD data is available over the study area from the ADL algorithm for the years 1983-2014 (Xue et al., 2017) and from ATSR-2 and AATSR using the ADV algorithm for the years 1995-2003 (ATSR-2) and 2002-2012 (AATSR), with some gaps as described in de Leeuw et al. (2018). The consistency between ATSR-2 and AATSR AOD is addressed in Sogacheva et al. (2018a) who show that no systematic differences occur over China. In the current work we also use MODIS/Terra C6.1 merged DTDB AOD for comparison. The validation of this data set over China by Sogacheva et al. (2018a), using all available AERONET stations, shows it's good quality, with a small positive bias (0.06) with respect to AERONET AOD. As an example, AOD maps over the study area are shown in Fig. 2, seasonal aggregated for the full years 2000-2011 when data from all three sensors/algorithms are available: AATSR (ADV v2.31, L2, resolution 10x10 km²), AVHRR (ADL, resolution 0.05 °x 0.05 °) and MODIS C6.1 Merged DBDT (L2, resolution 10x10 km²).

Figure 2 shows the similar AOD patterns derived from the three sensors/algorithms, with high AOD in the SE part of the study area and lower elsewhere. As indicated above, these high and low AOD areas are roughly separated by the Heihe-Tengchong line which separates the NCP from the Loess Plateau in Shanxi Province and the Inner Mongolia Plateau. In the SW of the study area, in Shanxi Province, we see an area with elevated AOD stretching from the NE to the SW, i.e. in the Guanzhong

basin where pollution transported by NE winds from the BTH area accumulates between the Qinling Mountains and the Loess Plateau and mixes with locally produced pollution. Comparison of the spatial distribution shows the similarity of the AOD distributions for AVHRR and MODIS while for AATSR the spatial distribution deviates with low AOD over Inner Mongolia, i.e. over the Gobi desert with high surface reflectance. ADV AOD retrieval is known to often be unsuccessful over bright areas and hence also aggregated AODs are too low (de Leeuw et al., 2018, Sogacheva et al., 2108a). AVHRR ADL appears to produce more credible AOD values in such conditions, as suggested by the similarity of the AOD patterns to those from MODIS. Below this statement will be put in perspective with the validation results from the ground-based reference data set. Quantitatively, Figure 2 shows that in the south of the study area, i.e. south of 41 N, the MODIS AOD is overall higher than that retrieved from AATSR which in turn is overall somewhat higher than that from AVHRR. The smoother MODIS AOD, likely due to the larger number of data points because of the larger swath, together with the higher AOD and the scales chosen to plot the AOD maps, results in larger variability and patterns which are not as clearly revealed by ATSR and AVHRR. North from 41° AATSR AOD retrievals are often not successful so a quantitative comparison cannot be made at these latitudes. As regards the comparison of the other two sensors north of 41 N MODIS is overall higher than AVHRR, except, for instance, over an area in Inner Mongolia just north of 41 N where the MODIS AOD is close to zero while AVHRR provides AOD values between 0.1 and 0.2.



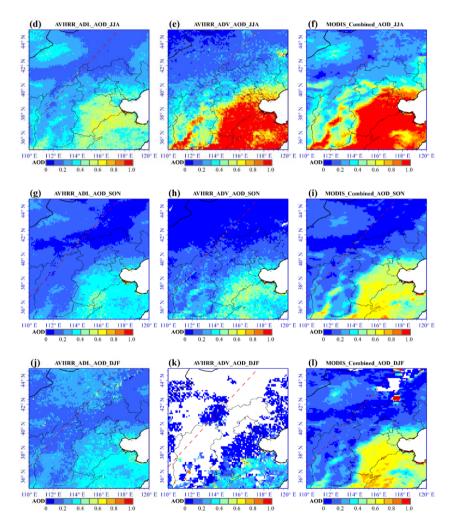


Figure 2: AOD over the study area retrieved from AATSR (ADV v2.31, L2, 10x10 km2), AVHRR (ADL, 0.05 ° x 0.05°) and MODIS C6.1 Merged DTDB (MODIS C6.1, L2, 10x10 km2), seasonal aggregated over the years 2000-2011.

Below the satellite AOD data will be validated versus the ground-based reference data from AERONET and CARSNET, for all stations available in the study area, as indicated above. The AVHRR data will also be compared with the radiation-derived AOD data available from the Beijing station which are especially useful for the earlier period when no other reference data are available. Next the satellite-derive AOD time series will be compared with the reference data to evaluate for each of the data sets when they are most useful. For AVHRR and MODIS, with wide swath, this will be done using monthly averaged AOD. For ATSR, with a much smaller swath, the data volume is too small for monthly averaging in a statistically meaningful sense and therefore seasonal averages will be used. In the direct comparison of monthly time series from other sensors, this may lead to an apparent shift in the AOD peak values.

3.2 Data quality: validation

0 3.2.1 Procedure

The MODIS and ATSR AOD data sets used in this study have been validated versus sun photometer reference data, on global and regional scales. With a focus on China, the ATSR v2.31 and MODIS C6.1 have been validated using AERONET data, for mainland China, for ten regions across China and for different seasons (de Leeuw et al., 2018, Sogacheva et al., 2018a). However, as shown in de Leeuw et al. (2018) and Sogacheva et al. (2018a), strong regional differences occur in both the seasonal and long-term AOD variations. Therefore, to achieve an unbiased comparison between the different data sets over the current study area, the MODIS C6.1 and ATSR data used in this study are validated versus the available reference data, together with the AVHRR data. The ADL data over the study area was earlier validated by Xue et al. (2017), but only AERONET stations were used while in the current study the complementary information (AOD) from CARSNET is also used.

Collocation of satellite and reference data is important for validation. Here we follow the spatio-temporal collocation method proposed by Ichoku et al., (2002), i.e. the satellite data were averaged over an area of 5 x 5 pixels (ca. 50 x 50km² at nadir) around the sun photometer location, whereas the sun photometer data were averaged over ±30 min around satellite overpass time. This spatio-temporal collocation method has been widely used for the validation of satellite aerosol products, for instance for MODIS (Ichoku et al., 2002, Chu et al., 2002, Remer et al., 2005, Levy et al., 2010, Sayer et al., 2014), AATSR (Che et al., 2016), and AVHRR (Riffler et al., 2010, Xue et al., 2017).

- In the data presentation and discussion expected error (EE) envelopes are used which apply to MODIS DT over land, i.e. expected errors of ±(0.05+0.15τ_{AERONET}) (Levy et al., 2013). For comparison, this value is also used for ATSR and AVHRR although the actual expected error envelopes for these sensors, which were not designed for aerosol retrieval, is expected to be higher than for MODIS. For ATSR a per-pixel uncertainty is provided rather than EE (Kolmonen et al., 2016) and for the AVHRR ADL (Xue et al., 2017) uncertainties have not been estimated.
- 30 The Beijing solar radiation station is located to the SW of the Beijing AERONET site at a distance of approximately 20km.
 AOD differences at these two sites are likely to occur due to emissions and secondary formation processes over this urban area,

but these have not been accounted for in the current study where collocations include averages over areas larger than the separation between these sites. Therefore, BEM AOD is directly compared to the sun photometer AOD as was done by Xu et al. (2015).

3.2.2 Results

20

5 Figure 3 shows scatterplots of AOD, for each of the three data sets individually, versus ground-based AOD reference data from all AERONET and CARSNET (3 years: 2007, 2008 and 2010) sites in the study area. Large differences occur between the three data sets. Starting with MODIS/Terra C6.1 merged DTDB, which has the largest number of collocations, the scatterplot in Fig. 3a shows the excellent performance for AOD up to 1.3 with the bin-averaged AOD less than 0.05 below the identity line and 69.5% of all data points within expected error of ±(0.05+0.15τ_{AERONET}). For AOD of 1.3 and larger the bin-averaged AOD values are much lower than the reference values, although still within one standard deviation. For AOD>2.6, MODIS does provide values but they are all well below the identity line. These results indicate that MODIS C6.1 AOD over the study area is reliable for AOD up to 1.3, but for higher AOD their use is not recommended.

ATSR with a swath width of 500 km provides much less collocations than MODIS but the large difference in performance (compare Fig. 3a and 3c) cannot be explained by the swath alone and is likely due to the failure of ADV to provide adequate retrievals over bright surfaces. Figure 3b shows that ADV provides AOD values up to 2.7 but for AOD > 1.3 the values are widely scattered around the identity line with large deviations from the sun photometer values. Also for lower AOD a systematic underestimation is observed which increases with the AOD value up to 0.8 where the ADV-retrieved AOD is about 0.3 low. For larger AOD, up to 1.3, ADV continues to underestimate the AOD with a similar amount. Although 52% of the data points are within the EE, ADV clearly underestimates the AOD over this area and the bin-averaged AOD follows the lower EE limit rather than the identity line, for AOD up to 1.0, leading to the conclusion that ADV AOD is about 0.15 x AOD low

AVHRR with a swath width of 2900 km is expected to provide the largest number of collocations but actually it is about 20% lower than for MODIS, likely due to failure of ADL to provide a valid retrieval in all situations. ADL does not provide retrievals for AERONET AOD > 2 and the data in Fig. 3a show the large scatter around the identity line and the large underestimation for AOD > 0.5. For AOD larger than about 0.8, the binned ADL AOD data deviate from the identity line by more than the expected error. For AOD up to 0.5, the bin-averaged values are within the EE (58.1% of the data), but systematically deviate from the identity line and underestimation increases with increasing AOD.

In conclusion, in the study area, MODIS provides reliable AOD for values up to 1.3 with a slight underestimation. This is in contrast with the findings of Sogacheva et al. (2018a) who observed a slight overestimation (bias 0.06) of the MODIS/Terra C6.1 merged DTDB AOD using all AERONET data available over China, but no CARSNET. CARSNET could provide more reference data where there are not AERONET data except Beijing region. Also for ATSR ADV v2.31 data the validation presented in de Leeuw et al. (2018), also using only AERONET data similar to Sogacheva et al. (2018a), deviates from that

presented here, with a slight underestimation resulting in a bias of 0.07. Clearly the selection of the smaller study area affects the validation results, with for MODIS slightly better results (smaller bias) than over all China, but for ATSR ADV the performance is less good. AVHRR AOD shows a large scatter and does not follow the identity line even for low values, underestimates for AOD larger than 0.5 and appears to fail for AOD larger than about 0.8.

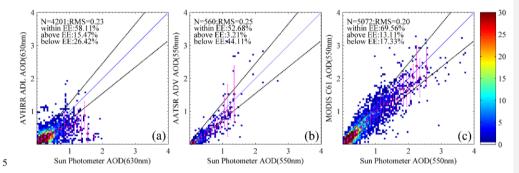


Figure 3: Scatter density plots of satellite-retrieved AOD versus sun photometer retrieved AOD data from AERONET (all available data) and CARSNET (3 years: 2007, 2008 and 2010) stations in the study area. The red symbols represent the averaged satellite-retrieved AOD binned in 0.1 sun photometer AOD intervals and the vertical lines on each circle represent the 1-sigma standard deviation of the fits. The colour bar on the right indicates the number of data points. Parameters for a LSQ fit to all data points are presented in the upper left corner, where N is the total number of collocated pairs and RMS is the root mean square error. The blue line represents the identity line and the black dotted lines represent the MODIS EE.

Table2. Metrics obtained from the evaluation of three AOD products. N is number of collocated data points, MSA is mean satellite retrieved AOD, MAA is mean sun photometer (AERONET and CARSNET) derived AOD, RMB is the ratio MAA/MSA. MBE is mean of AOD bias given by $\tau_{sat} - \tau_{aero}$. R is the Pearson correlation coefficient for a LSQ fit to all data points. RMS is the root mean square error. EE is the expected error, EE_a is the fraction above the EE, EE_b is the fraction below the EE.

Product	Network	N	MSA	MAA	RMB	MBE	RMS	R	EE	EE_a	EE_b
MODIS	AERO&CARS	5072	0.441	0.464	0.950	-0.023	0.204	0.910	69.56%	13%.11	17.33%
ADV	AERO&CARS	560	0.289	0.396	0.729	-0.107	0.252	0.843	52.68%	3.21%	44.11%
AVHRR	AERO&CARS	4201	0.237	0.296	0.802	-0.059	0.229	0.602	58.11%	15.47%	26.42%

20 Seasonal scatterplots are presented in the Appendix and discussed in the context of the seasonal variations observed in the AOD time series presented below.

3.3 Comparison of satellite-derived AOD time series with reference data

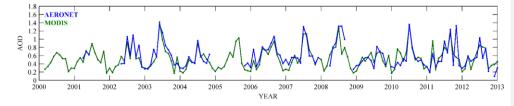
In this section time series of satellite-retrieved AOD data will be compared with time series available from the AERONET sites in Beijing and XiangHe, for which the longest time series are available, and with AOD derived from broadband radiances measured at the Beijing radiation station.

5 3.3.1 AERONET AOD in Beijing

AOD time series for the period 1997-2012, retrieved from AVHRR, ATSR and MODIS data and spatially collocated with the Beijing AERONET site, are presented in Fig. 4 together with monthly-averaged AERONET AOD for reference. The time series presented here start in 2000 because for the earlier years no AERONET and MODIS data are available. The data in Fig. 4 show that AVHRR is almost always lower than AERONET and that the high AOD values obtained from AERONET, usually in the summer, are not reproduced by AVHRR. This is not unexpected since the scatterplots in Fig. 3 show that AVHRR fails to retrieve high AOD. For these individual data points hardly any values larger than 1 were retrieved and hence the monthly averages in Fig. 4 are lower than that. Figure 4 shows that the highest AVHRR AOD values of about 0.8 occur in 2002 and 2003 and thereafter do not exceed 0.6. The monthly averaged AERONET AOD peak values occur mostly in the summer and are about 1.4. However, in other seasons the differences are much smaller and the two time series seem to trace rather well with an offset of about 0.1. In particular in the winter AVHRR AOD is close to the reference value. These seasonal differences are confirmed by the scatterplots presented in the Appendix. The results lead to the conclusion that, for the Beijing site, for low AOD the ADL algorithm provides quite reasonable results but for high AOD improvement is needed to provide reliable time series.

The MODIS data in Fig. 4 (top) trace the AERONET AOD very well, both in the summer and in other seasons, with MODIS AOD often just a little smaller as can also be observed from the scatterplots in the Appendix. This shows that MODIS serves as a good reference for the situation encountered at the Beijing site.

This does not apply to the ATSR AOD. Although ATSR AOD variations follow those from AERONET, and even reproduce the high summer values in some years (2007 and 2011), the ATSR values are most of the time smaller and especially in the winter the ATSR AOD is much too low. As discussed before, this may be due to the failure of ADV to produce a valid retrieval over bright surfaces.



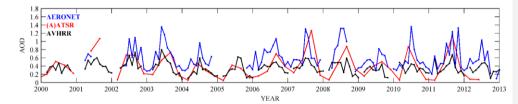


Figure 4: AOD time series for MODIS (top), (A)ATSR and AVHRR (bottom), with AERONET AOD (blue) as reference, over the Beijing site for the years 2000-2012. For (A)ATSR seasonally averaged AOD is plotted, for AVHRR and MODIS the AOD data are monthly averages.

5 3.3.2 AERONET AOD in XiangHe

Figure 5 shows a comparison of the AOD time series over the XiangHe AERONET site, similar to that over Beijing in Fig. 4. It is noted that, for comparison, in both cases the time series are plotted for the years 2000-2012, but that the XiangHe time series start at the end of 2004 (except for a few data points in 2001), i.e. later than in Beijing. Comparison of the AERONET time series shows that over XiangHe the AOD in the summer is somewhat higher and peaks often in the same years as over Beijing, but there are also differences such as in 2008 when the AOD in XiangHe reached the maximum monthly-averaged value of 1.5. As in Beijing, the AVHRR retrieval did not reproduce these high values in the summer, but for the other months the AVHRR/AERONET comparison is better than in Beijing and, with some exceptions, the AOD data trace very well. The seasonal scatter density plots in the Appendix, Fig. A3, confirm that no high AOD is retrieved in the summer, as opposed to other seasons. These scatterplots also confirm the underestimation of ADL, by about one EE, in each of the seasons except in the winter where ADL performance compares better to AERONET.

In contrast, the ATSR retrieval algorithm does reproduce the higher values, and in some years the seasonally averaged AOD compares favourably with the AERONET AOD (e.g. in 2007 and 2011), but in other years, such as 2008 and 2010 the maximum AOD is not observed by AATSR, which in part may be due to the seasonal averaging. Also, as over Beijing, ATSR retrieval does not work well in the winter time, possibly due to the high surface reflectance in the dry season, and the seasonal AOD is much lower than AERONET.

For MODIS the AOD compares well with AERONET but not as good as over Beijing. During some periods, e.g. in 2005 and during 2008-2011, MODIS is too high. This is also shown by the validation results in the Appendix. The reason for this difference between Beijing and XiangHe, which is located close to Beijing to the ESE (see Fig. 1), has not been further investigated. Possibly, the surface properties, urban and build-up for Beijing versus cropland for XiangHe, may affect the retrieval results as was also indicated in de Leeuw et al. (2018), but for MODIS C6.0.

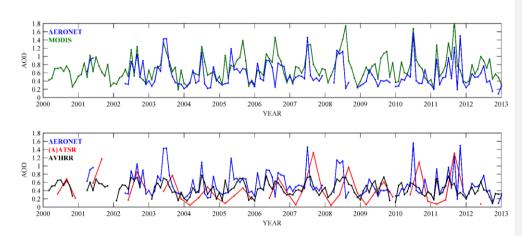


Figure 5: AOD time series for MODIS (top), (A)ATSR and AVHRR (bottom), with AERONET AOD (blue) as reference, over the XiangHe site for the years 2000-2012. For ATSR seasonally averaged AOD is plotted, for AVHRR and MODIS the AOD data are monthly averages.

3.3.3 Broadband extinction method (BEM) AOD in Beijing

Figure 6 shows the comparison of the monthly-averaged AOD time series from AVHRR with those derived from BEM data, as well as a comparison of the BEM AOD with AERONET and with MODIS, as reference data. Monthly radiance data at the Beijing site have been validated by Xu et al. (2015) for the years 2002-2012 using both MODIS C5 L3 AOD and AERONET L2 data (version 2). In the current study MODIS C6.1 and AERONET version 3 are used, for the years 2000-2012, and the results from the comparison of the BEM AOD with these data sets, shown in Fig. 7 (left), are similar to those presented in Fig. 2 of Xu et al. (2015). Also the time series in Fig. 6 show very good comparisons between the BEM and MODIS and AERONET AOD. Hence the BEM AOD data provide another reference data set for the AVHRR-retrieved AOD which can be used for the period 1987-2012 as presented in Fig. 8. Figure 8 shows that in this earlier period (1987-2000) the AOD was generally lower and the monthly averaged AOD compares better with BEM AOD than for the later period 2000-2012, when AOD was higher during especially the summer months. This is consistent with in right scatter in fig. 7 that almost all of AVHRR-BEM matchups are below identify line when AOD larger than 0.6. The overall conclusion remains the same, however, that the AVHRR retrieval needs improvement for high AOD and only values up to about 0.6 can be used.

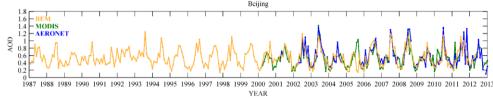


Figure 6: AOD time series for BEM (orange), with MODIS(green) and AERONET AOD (blue) as reference, over the Beijing site for the years 1987-2012.

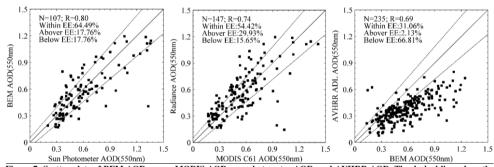


Figure 7: Scatter plots of BEM AOD versus MODIS AOD, sun photometer AOD, and AVHRR AOD. The dashed lines show the MODIS expected error of $\pm (0.05+0.15\tau_{AERONET})$.

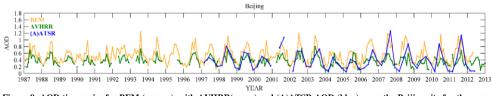


Figure 8: AOD time series for BEM (orange), with AVHRR(green) and (A)ATSR AOD (blue), over the Beijing site for the years 1987-2012.

3.3 Seasonal comparison of satellite-derived AOD

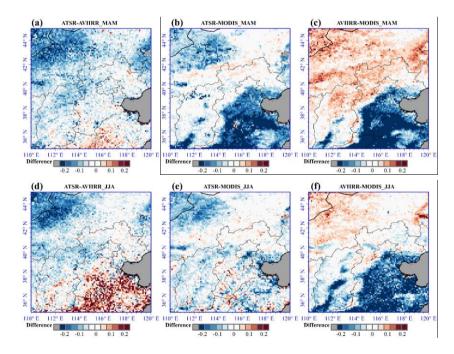
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The above results and those from Sogacheva (2018a) and de Leeuw (2018) show MODIS data are with high quality over China. Therefore, we select MODIS as reference data both on time series and spatial coverage. Last session has compared satellite-retrieved data with ground-based data on time series, this session will compare ATSR and AVHRR data with MODIS data spatially.

As Fig. 2 shows, MODIS could provide AOD higher than 0.7 in SE part where ATSR and AVHRR data are with close pattern that hardly AOD higher than 0.4. Hence, in spring the SE part in Fig. 9b&c present same AOD difference that lower than

MODIS more 0.2 and Fig. 9a present no obvious difference which is AOD difference of ATSR minus AVHRR. When the season is changed to summer time, AOD in SE part is increasing higher than 0.8 even 1.0, ATSR and MODIS could capture high AOD well but AVHRR cannot. This leads Fig. 9f presents dark blue in SE part. The situation in autumn is quite like it in spring when AOD in SE part is decreasing lower than 0.7, the patterns of ATSR and AVHRR are quite similar in SE which leads AOD difference in Fig. 9g&h are quite similar. Due to ability of ADV in winter, ATSR miss retrievals approximately in half of areas but remain part are close to AVHRR and MODIS as Fig. 9j&k show.

As discussed above, there are not enough successful retrievals at north of 41°, so we don't discuss AOD difference over part at north from 41°, otherwise AVHRR follows MODIS well in this part with slight overestimation lower than 0.1. This mainly because AVHRR has longer spectral band leading to less sensitivity to low AOD. In addition, ATSR data sample volume is much lower than other two due to sensor swath and algorithm ability, so seasonal averaged AOD are not smooth as other two.



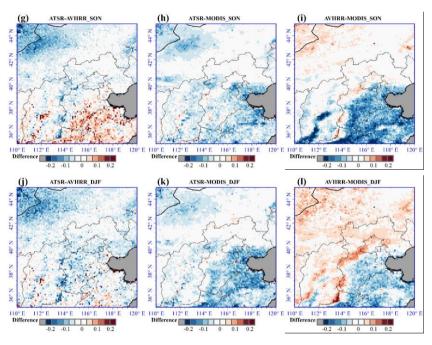


Figure 9: AOD difference over the study area retrieved from AATSR (ADV v2.31, L2, 10x10 km²), AVHRR (ADL, 0.05 ° x 0.05°) and MODIS C6.1 Merged DTDB (MODIS C6.1, L2, 10x10 km²), seasonal aggregated over the years 2000-2011.

4 Discussion and conclusion

The validation exercises and time series comparisons presented in Sect. 3 show the stronger and weaker points of the ATSR-and AVHRR-retrieved AOD Climate Data Records (CDRs), with AVHRR being better in the winter and ATSR better in the summer. These differences are also very clear in the direct comparison of the AVHRR and ATSR time series, shown in Fig. 10 for the Beijing site and in Fig. 11 for the XiangHe site. Figure 10 also shows that before 2007 these time series trace quite well and could be combined into a single time series while taking into account the validation results for that period only. In particular the use of the ATSR winter AOD data will have to be considered carefully and may possible have to be given little weight. However, after 2007 large differences are observed with quite high ATSR AOD in the summer while that retrieved from AVHRR has a maximum value of about 0.6. The comparison with MODIS shows that the ATSR AOD values are credible and follow those of MODIS (apart from the shift due the use of seasonal ATSR vs monthly MODIS AOD averages), whereas

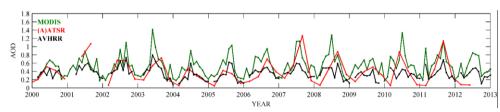
the AVHRR AOD is much too low. In the winter the opposite is observed: ATSR is too low with values close to zero while AVHRR traces MODIS very well. The intercomparison of the ATSR and AVHRR time series for XiangHe in Fig. 11 shows a similar pattern as for Beijing, with a good comparison before 2007 and a larger summer/winter difference in the ATSR data there-after, resulting in increased discrepancy between the ATSR and AVHRR data sets. However, the comparison with MODIS shows an offset between ATSR and MODIS in the years 2004-2007 which seems to be due to larger overestimation of the MODIS AOD at the XiangHe site than at the Beijing site. Also in some other years the ATSR/MODIS difference appears to be higher in XiangHe than in Beijing, due to higher MODIS AOD. The comparison between AERONET and MODIS AOD data in Figures 4 and 5 show that the difference between the AOD measured at both sites is smaller for AERONET than that for MODIS and hence this may be a MODIS retrieval issue.

10 As the statistics marked in A2 and A3, the data volume of collocated matchups with ground-based reference data may lead to biased validation results and time series comparison. Therefore, pixel by pixel comparison with reference data is necessary. The seasonal averaged AOD and AOD difference maps provides information on not only seasonal but also pixel by pixel comparison. As reference data, MODIS AOD could be not only sensitive to low AOD but also used to detect high AOD that it performs best in different seasons (see A1). As discussed above, AVHRR failed to retrieve AOD over 0.6 for most cases leading to big differences with ATSR and MODIS in the summer in the SE (of study area). However, the AOD patterns of ATSR and AVHRR in other seasons are similar. ATSR has ability to retrieve high AOD, otherwise its data coverage limits its performance as Fig. 9 and c shows that difference between ATSR and MODIS AOD is close to zero while the difference between AVHRR and MODIS AOD is lower than -0.2 for most of area of the SE. This is consistent with last conclusion.

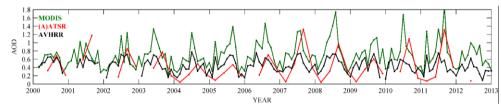
BEM AOD data set could not only provide consistent AOD from 1980s to the current but also be with credible data quality compared with MODIS and AERONET AOD. As shown in Fig. 8, BEM AOD agrees MODIS and AERONET AOD very well with close pattern at overlaying years. Above, it is concluded that AVHRR AOD agrees ATSR well, however there is no ATSR AOD when it goes back to early 1990s and 1980s, AVHRR is the only choice to extend ATSR AOD data set. Still, AVHRR AOD missed most of high AOD over 0.6 in the summer but follows BEM AOD well in other seasons and has similar annual patterns. Only one BEM station weaker conclusion of this paper, however there are at least ten BEM stations in whole China region as mentions in Xu et al. (2015), hence it is expected to extend AVHRR data set to whole China (which is our current focus).

In conclusion, the possible combination of the AVHRR and ATSR CDRs, possibly also including MODIS, into a multi-decadal time series is not straightforward. Although MODIS performance is better that that of ATSR and AVHRR, the latter two sensors go back in time before MODIS and hence provide a unique source of information. In addition, none of the satellite sensors performs better than the others at any time and at any location and their combination taking into account the strengths and weaknesses of each of them, may result in a more significant CDR than each of them individually as indicated by that AVHRR failed to retrieve AOD over 0.6 while ATSR could and ATSR AOD is not credible from 41° to the north border while AVHRR AOD is acceptable. Furthermore, the comparison shows that before 2007 the AVHRR and ATSR compare quite well

and could be used for a meaningful extension of the AOD data records to the early 1980s. However, this conclusion does not apply to the northern sub-region where the surface conditions seem unfavourable for AOD retrieval from ATSR using ADV.



5 Figure 10: Comparison of AOD time series over the Beijing AERONET site retrieved from the satellite sensors AVHRR, MODIS and (A)ATSR, for the overlapping years 2000-2015. Note that for AVHRR and MODIS monthly mean AOD data are plotted, while for (A)ATSR they are seasonal means which may provide a shift in the extreme values.



0 Figure 11: As Fig. 10, over the XiangHe site.

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References

- Allen, R.C., Durkee, P.A., and Wash, C.H.: Snow/Cloud Discrimination with Multispectral Satellite Measurements, J. Appl. Meteorol. Clim., 29, 994-1004, doi: 10.1175/1520-0450(1990)029<0994:SDWMSM>2.0.CO;2, 1990.
- Alizadeh-Choobari, O., Sturman, A., and Zawar-Reza, P.: A global satellite view of the seasonal distribution of mineral dust and its correlation with atmospheric circulation, Dynam. Atmos. Oceans, 68, 20–34, doi: 10.1016/j.dynatmoce.2014.07.002, 2018.
 - Ångström, A.: Solar and terrestrial radiation. Report to the international commission for solar research on actinometric investigations of solar and atmospheric radiation, Q. J. Roy. Meteor. Soc., 50, 121-126, doi:10.1002/qj.49705021008, 1924.
- Bai, J., de Leeuw, G., van der A, R., De Smedt, I., Theys, N., Van Roozendael, M., Sogacheva, L., and Chaia, W.: Variations and photochemical transformations of atmospheric constituents in North China, Atmos. Environ., 189, 213-226, https://doi.org/10.1016/j.atmosenv.2018.07.004, 2018.
 - Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B., and Zhang, X.: Clouds and Aerosols. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.
 - Che, H., Zhang, X. Y., Chen, H. B., Damiri, B., Goloub, P., Li, Z. Q., Zhang, X. C., Wei, Y., Zhou, H. G., Dong, F., Li, D. P., and Zhou, T. M.: Instrument calibration and aerosol optical depth validation of the China Aerosol Remote Sensing Network, J. Geophys. Res., 114, D03206, doi:10.1029/2008JD011030, 2009.
- 20 Che, H., Zhang, X.-Y., Xia, X., Goloub, P., Holben, B., Zhao, H., Wang, Y., Zhang, X.-C., Wang, H., Blarel, L., Damiri, B., Zhang, R., Deng, X., Ma, Y., Wang, T., Geng, F., Qi, B., Zhu, J., Yu, J., Chen, Q., and Shi, G.: Ground-based aerosol climatology of China: aerosol optical depths from the China Aerosol Remote Sensing Network (CARSNET) 2002–2013, Atmos. Chem. Phys., 15, 7619–7652, doi: 10.5194/acpd-15-12715-2015, 2015.
- Che, Y., Xue, Y., Mei, L., Guang, J., She, L., Guo, J., Hu, Y., Xu, H., He, X., Di, A., and Fan, C.: Technical note:

 Intercomparison of three AATSR Level 2 (L2) AOD products over China, Atmos. Chem. Phys., 16, 9655–9674, doi: 10.5194/acp-16-9655-2016, 2016.
 - Chu, D. A., Kaufman, Y. J., Ichoku, C., Remer, L. A., Tanre , D., and Holben, B. N.: Validation of MODIS aerosol optical depth retrieval over land, dust, Geophys, Res. Lett., 29, MOD2-1–MOD2-4, doi: 10.1029/2001GL013205, 2002.
- de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.:

 Production flux of sea spray aerosol, Rev. Geophys., 49, doi: 10.1029/2010RG000349, 2011.
- de Leeuw, G., Holzer-Popp, T., Bevan, S., Davies, W. H., Descloitres, J., Grainger, R. G., Griesfeller, J., Heckel, A., Kinne, S., Klüser, L., Kolmonen, P., Litvinov, P., Martynenko, D., North, P., Ovigneur, B., Pascal, N., Poulsen, C., Ramon, D., Schulz, M., Siddans, R., Sogacheva, L., Tanr & D., Thomas, G. E., Virtanen, T. H., von Hoyningen-Huene, W., Vountas,

- M., and Pinnock, S.: Evaluation of seven European aerosol optical depth retrieval algorithms for climate analysis, Remote Sens. Environ., 162, 295-315, doi:10.1016/j.rse.2013.04.023, 2015.
- de Leeuw, G., Sogacheva, L., Rodriguez, E., Kourtidis, K., Georgoulias, A. K., Alexandri, G., Amiridis, V., Proestakis, E., Marinou, E., Xue, Y., van der A, R.: Two decades of satellite observations of AOD over mainland China using ATSR-2,
- 5 AATSR and MODIS/Terra: data set evaluation and large-scale patterns, Atmos. Chem. Phys., 18, 1573-1592, doi: 10.5194/acp-18-1573-2018, 2018.
 - Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, J. Geophys. Res.-Atmos., 104, 31333–31349, doi: 10.1029/1999JD900923, 1999.
- 10 Flowerdew, R. J., and Haigh, J. D.: An approximation to improve accuracy in the derivation of surface reflectance from multi-look satellite radiometers, Geophys. Res. Lett., 23, 1693–1696, doi: 10.1029/95GL01662, 1995.

- Giles, David M., Sinyuk, Alexander, Sorokin, Mikhail G., Schafer, Joel S., Smirnov, Alexander, Slutsker, Ilya, Eck, Thomas F., Holben, Brent N., Lewis, Jasper R., Campbell, James R., Welton, Ellsworth J., Korkin, Sergey V., Lyapustin, Alexei I.: Advancements in the Aerosol Robotic Network (AERONET) Version 3 database automated near-real-time quality control algorithm with improved cloud screening for Sun photometer aerosol optical depth (AOD) measurements, Atmos. Meas. Tech., 12, 169–209, doi: 10.5194/amt-12-169-2019, 2019.
- Govaerts, Y.M., Wagner, S., Lattanzio, A., and Watts P.: Joint retrieval of surface reflectance and aerosol optical depth from MSG/SEVIRI observations with an optimal estimation approach: 1. Theory, J. Geophys. Res.-Atmos., 115, D02203, doi:10.1029/2009JD011779, 2010.
- 20 Guerreiro, C.B.B., Foltescu, V., and de Leeuw, F.: Air quality status and trends in Europe, Atmos. Environ., 98, 376–384, doi: 10.1016/j.atmosenv.2014.09.017, 2014.
 - Guo, J.P., Zhai, P., Wu, L., Cribb, M., Li, Z., Ma, Z., Wang, F., Chu, D., Wang, P., and Zhang, J.: Precipitation and air pollution at mountain and plain stations in northern China: Insights gained from observations and modeling, J. Geophys. Res. -Atmos., 119 (8), 4793-4807, doi: 10.1002/2013jd021161 2014.
- 25 Guo, J., Deng, M., Lee, S. S., Wang, F., Li, Z., Zhai, P., Liu, H., Lv, W., Yao, W., and Li, X.: Delaying precipitation and lightning by air pollution over the Pearl River Delta. Part I: Observational analyses, J. Geophys. Res. Atmos., 121, 6472–6488, doi: 10.1002/2015JD023257, 2016a.
 - Guo, M., Sun, L., and Xu, X.: Inter-comparison of aerosol optical thickness from MODIS, MISR, and OMI using measurements from solar radiation stations in China, in: Proceedings of SPIE 9876, Remote Sensing of the Atmosphere, Clouds, and Precipitation VI, New Delhi, India, 6 September 2016, doi:10.1117/12.2228000, 2016b.
 - Guo, J., Liu, H., Li, Z., Rosenfeld, D., Jiang, M., Xu, W., Jiang, J. H., He, J., Chen, D., Min, M., and Zhai, P.: Aerosol-induced changes in the vertical structure of precipitation: a perspective of TRMM precipitation radar, Atmos. Chem. Phys., 18, 13329-13343, https://doi.org/10.5194/acp-18-13329-2018, 2018.

- Hao, J., Wang, S., Liu, B., and He., K.: Designation of acid rain and SO2 control zones and control policies in China, J. Environ. Sci. Heal A, 35, 1901–1914, doi: 10.1080/10934520009377085, 2000.
- Hollmann, R., Merchant, C. J., Saunders, R., Downy, C., Buchwitz, M., Cazenave, A., Chuvieco, E., Defourny, P., de Leeuw, G., Forsberg, R., Holzer-Popp, T., Paul, F., Sandven, S., Sathyendranath, S., van Roozendael, M., and Wagner, W.: The ESA climate change initiative: satellite data records for essential climate variables, B. Am. Meteorol. Soc., 94, 1541-1552, doi: 10.1175/BAMS-D-11-00254.1, 2013.

10

- Holzer-Popp, T., de Leeuw, G., Griesfeller, J., Martynenko, D., Klüser, L., Bevan, S., Davies, W., Ducos, F., Deuz é, J. L., Graigner, R. G., Heckel, A., von Hoyningen-Hüne, W., Kolmonen, P., Litvinov, P., North, P., Poulsen, C. A., Ramon, D., Siddans, R., Sogacheva, L., Tanre, D., Thomas, G. E., Vountas, M., Descloitres, J., Griesfeller, J., Kinne, S., Schulz, M., and Pinnock, S.: Aerosol retrieval experiments in the ESA Aerosol_cci project, Atmos. Meas. Tech., 6, 1919-1957, doi:10.5194/amt-6-1919-2013, 2013.
- Hsu, N. C., Tsay, S. -C., King, M. D., and Herman, J. R.: Aerosol properties over bright-reflecting source regions, IEEE T. Geosci. Remote, 42, 557-569, doi: 10.1109/TGRS.2004.824067, 2004.
- Hsu, N. C., Jeong, M.-J., Bettenhausen, C., Sayer, A. M., Hansell, R., Seftor, C. S., Huang, J., and Tsay, S.-C.: Enhanced Deep

 Blue aerosol retrieval algorithm: The second generation, J. Geophys. Res. Atmos., 118, 9296–9315,

 doi:10.1002/jgrd.50712, 2013.
 - Hsu, N. C., Lee, J., Sayer, A. M., Carletta, N., Chen, S.-H., Tucker, C. J., Holben, B. N., and Tsay, S.-C.: Retrieving Near-Global Aerosol Loading over Land and Ocean from AVHRR, J. Geophys. Res. Atmos., 122, doi:10.1002/2017JD026932, 2017.
- 20 Ichoku, C, Chu, D. A., Mattoo, S., Kaufman, Y. J., Remer, L. A., Tanre ´, D., Slutsker, I., and Holben, B. N.: A spatio-temporal approach for global validation and analysis of MODIS aerosol products, Geophys. Res. Lett., 29, MOD1-1-MOD1-4, doi: 10.1029/2001GL013206, 2002.
 - Jin, Y., Andersson, H., and Zhang, S.: Air Pollution Control Policies in China: A Retrospective and Prospects, Int. J. Environ. Res. Public Health, 13, 1219, doi:10.3390/ijerph13121219, 2016.
- 25 Kang, H., Zhu, B., van der A. R., Zhu, C., de Leeuw, G., Hou, X., and Gao, J.: Natural and anthropogenic contributions to long-term variations of SO2, NO2, CO, and AOD over East China, Atmos. Environ., 215, 284-293, https://doi.org/10.1016/j.atmosres.2018.09.012, 2018.
 - Kaufman, Y. J., Wald, A. E., Remer, L. A., Gao, B. C., Li, R. R., and Flynn, L.: The MODIS 2.1-mu m channel Correlation with visible reflectance for use in remote sensing of aerosol, IEEE T. GEOSCI. REMOTE, 35, 1286-1298, doi: 10.1109/36.628795, 1997.
 - Koren, I., Dagan, G., and Altaratz, O.: From aerosol-limited to invigoration of warm convective clouds, Science, 344(6188), 1143-1146, 2014.

- Kolmonen, P., Sogacheva, L., Virtanen, T.H., de Leeuw, G., and Kulmala, M.: The ADV/ASV AATSR aerosol retrieval algorithm: current status and presentation of a full-mission AOD data set, Int. J. Digit. Earth, 9:6, 545-561, doi: 10.1080/17538947.2015.1111450, 2016.
- Levy, R. C., Remer, L. A., Kleidman, R. G., Mattoo, S., Ichoku, C., Kahn, R., and Eck, T. F.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmos. Chem. Phys. 10, 10399-10420, doi:10.5194/acp-10-10399-2010, 2010.
 - Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The Collection 6 MODIS aerosol products over land and ocean, Atmos. Meas. Tech., 6, 2989-3034, doi:10.5194/amt-6-2989-2013, 2013.
- Levy, R. C., Munchak, L. A., Mattoo, S., Patadia, F., Remer, L. A., and Holz, R. E.: Towards a long-term global aerosol optical depth record: applying a consistent aerosol retrieval algorithm to MODIS and VIIRS-observed reflectance, Atmos. Meas. Tech., 8, 4083-4110, https://doi.org/10.5194/amt-8-4083-2015, 2015.
 - Li, J., Liu, R., Liu, S., Shiu, C., Wang, J., and Zhang, Y.: Trends in aerosol optical depth in northern China retrieved from sunshine duration data, Geophys. Res. Lett., 43, 431-439, doi: 10.1002/2015GL067111, 2016.
 - Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu B.: Aerosol and boundary-layer interactions and impact on air quality. Natl. Sci. Rev., 4, 810–833. doi: 10.1093/nsr/nwx117, 2017a.

20

- Li, Z., Xu, H., Li, K. T., Li, D. H., Xie, Y. S., Li, L., Zhang, Y., Gu, X. F., Zhao, W., Tian, Q. J., Deng, R. R., Su, X. L., Huang, B., Qiao, Y. L., Cui, W. Y, Hu, Y., Gong, C. L., Wang, Y. Q., Wang, X. F., Wang, J. P., Du, W. B., Pan, Z. Q., Li, Z. Z., and Bu, D.: Comprehensive study of optical, physical, chemical and radiative properties of total columnar atmospheric aerosols over China: An overview of Sun-sky radiometer Observation NETwork (SONET) measurements, Bull. Amer. Meteor. Soc., doi:10.1175/BAMS-D-17-0133.1, 2018.
- Miao, Y., Guo, J., Liu, S., Liu, H., Zhang, G., Yan, Y., and He, J.: Relay transport of aerosols to Beijing-Tianjin-Hebei region by multi-scale atmospheric circulations, Atmos. Environ., 165, 35-45, doi: 10.1016/j.atmosenv.2017.06.032, 2017.
- Popp, T., de Leeuw, G., Bingen, C., Brühl, C., Capelle, V., Chedin, A., Clarisse, L., Dubovik, O., Grainger, R., Griesfeller, J.,
 Heckel, A., Kinne, S., Klüser, L., Kosmale, M., Kolmonen, P., Lelli, L., Litvinov, P., Mei, L., North, P., Pinnock, S.,
 Povey, A., Robert, C., Schulz, M., Sogacheva, L., Stebel, K., Stein Zweers, D., Thomas, G., Tilstra, L.G., Vandenbussche,
 S., Veefkind, P., Vountas, M., and Xue, Y.: Development, production and evaluation of aerosol Climate Data Records
 from European satellite observations (Aerosol_cci), Remote Sens., 8, 421, doi:10.3390/rs8050421, 2016.
 - Qiu, J.: A Method to Determine Atmospheric Aerosol Optical Depth Using Total Direct Solar Radiation, J. Atmos. Sci., 55, 744-757, doi: 10.1175/1520-0469(1998)055<0744:AMTDAA>2.0.CO;2, 1998.
 - Remer, L. A., Kaufman, Y. J., Tanré, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R.-R., Ichoku, C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B. N.: The MODIS aerosol algorithm, products, and validation, J. Atmos. Sci., 62, 947–973, doi: http://dx.doi.org/10.1175/JAS3385.1, 2005.

- Riffler, M., Popp, C., Hauser, A., Fontana, F., and Wunderle, S.: Validation of a modified AVHRR aerosol optical depth retrieval algorithm over Central Europe, Atmos. Meas. Tech., 3, 1255-1270, 2010.
- Robel, J., and Graumann, A.: NOAA KLM USER'S GUIDE with NOAA-N, N Prime, and MetOp SUPPLEMEN: https://www.nasa.gov/pdf/111742main noaa n booklet.pdf, 2014.
- 5 Salomonson, V. V., Barnes, W. L., Maymon, P. W., Montgomery, H. E., Ostrow, H.: MODIS Advanced facility instrument for studies of the earth as a system, IEEE T. Geosci. Remote, 27, 145-153, doi: 10.1109/36.20292, 1989.
 - Sayer, A. M., Hsu, N. C., Bettenhausen, C., and Jeong, M.-J.: Validation and uncertainty estimates for MODIS Collection 6 "Deep Blue" aerosol data, J. Geophys. Res. Atmos., 118, 7864–7872, doi:10.1002/jgrd.50600, 2013.
- Sayer, A. M., Munchak, L. A., Hsu, N. C., Levy, R. C., Bettenhausen, C., and Jeong, M.-J.: MODIS Collection 6 aerosol products: Comparison between Aqua's e-Deep Blue, Dark Target, and "merged" data sets, and usage recommendations, J. Geophys. Res. Atmos., 119, 13965–13989, doi: 10.1002/2014JD022453, 2014.
 - Sayer, A. M., Hsu, N. C., Bettenhausen, C., Jeong, M.-J., and Meister, G.: Effect of MODIS Terra radiometric calibration improvements on Collection 6 Deep Blue aerosol products: Validation and Terra/Aqua consistency, J. Geophys. Res. Atmos., 120, 12157-12174, doi: 10.1002/2015JD023878, 2015.
- Sayer, A. M., Hsu, N. C., Lee, J., Carletta, N., Chen, S. -H., and Smirnov, A.: Evaluation of NASA Deep Blue/SOAR aerosol retrieval algorithms applied to AVHRR measurements, J. Geophys. Res. Atmos., doi: 10.1002/2017JD026934, doi:10.1002/2017JD026934, 2017.
 - Seinfeld, J. H., Pandis, S. N., and Noone, K.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Phys. Today, 51, 88, doi: 10.1063/1.882420, 1998.
- 20 Sogacheva, L., Kolmonen, P., Virtanen, T. H., Rodriguez, E., Saponaro, G., and de Leeuw, G.: Post-processing to remove residual clouds from aerosol optical depth retrieved using the Advanced Along Track Scanning Radiometer, Atmos. Meas. Tech., 10, 491-505, doi:10.5194/amt-10-491-2017, 2017.
 - Sogacheva, L., de Leeuw, G., Rodriguez, E., Kolmonen, P., Georgoulias, A. K., Alexandri, G., Kourtidis, K., Proestakis, E., Marinou, E., Amiridis, V., Xue, Y., and van der A, R. J.: Spatial and seasonal variations of aerosols over China from two decades of multi-satellite observations Part 1: ATSR (1995–2011) and MODIS C6.1 (2000–2017), Atmos. Chem. Phys., 18, 11389-11407, https://doi.org/10.5194/acp-18-11389-2018, 2018a.

- Sogacheva, L., Rodriguez, E., Kolmonen, P., Virtanen, T. H., Saponaro, G., de Leeuw, G., Georgoulias, A. K., Alexandri, G., Kourtidis, K., and van der A, R. J.: Spatial and seasonal variations of aerosols over China from two decades of multisatellite observations. Part II: AOD time series for 1995–2017 combined from ATSR ADV and MODIS C6.1 for AOD tendencies estimation, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-288, in review, 2018b.
- Stark, M. S., Harrison, J. T. H., Anastasi, C.: The formation of nitrogen oxides by electrical discharges and implications for atmospheric lightning, J. Geophys. Res.-Atmos., 101, 6963-6969, doi: 10.1029/95JD03008, 1996.

- Stowe, L. L., McClain, E. P., Carey, R., Pellegrino, P., Gutman, G. G., Davis, P., Long, C., and Hart, S.: Global distribution of cloud cover derived from NOAA/AVHRR operational satellite data, Adv. Space Res., 11, 51–54, doi: 10.1016/0273-1177(91)90402-6, 1991.
- Stowe, L.L., Davis, P.A., and McClain, E.P.: Scientific basis and initial evaluation of the CLAVR-1 global clear cloud classification algorithm for the advanced very high resolution radiometer, J. Atmos. Ocean Technol., 16, 656–681, https://doi.org/10.1175/1520-0426(1999)016<0656:SBAIEO>2.0.CO;2, 1999.
 - Sundstr öm, A.-M., Kolmonen, P., Sogacheva L., and de Leeuw, G.: Aerosol retrievals over China with the AATSR Dual-View Algorithm, Remote Sens. Environ., 116, 189-198, doi: 10.1016/j.rse.2011.04.041, 2012.
- Tanré, D., Kaufman, Y. J., Herman, M., and Mattoo, S.: Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances, J. Geophys. Res. Atmos., 102, 16971-16988, doi: 10.1029/96JD03437, 1997.

- Theys, N., Campion, R., Clarisse, L., Brenot, H., van Gent, J., Dils, B., Corradini, S., Merucci, L., Coheur, P.-F., Van Roozendael, M., Hurtmans, D., Clerbaux, C., Tait, S., and Ferrucci, F.: Volcanic SO2 fluxes derived from satellite data: a survey using OMI, GOME-2, IASI and MODIS, Atmos. Chem. Phys., 13, 5945-5968, https://doi.org/10.5194/acp-13-5945-2013, 2013.
- 15 van der A, R. J., Mijling, B., Ding, J., Koukouli, M. E., Liu, F., Li, Q., Mao, H., and Theys, N.: Cleaning up the air: effectiveness of air quality policy for SO2 and NOx emissions in China, Atmos. Chem. Phys., 17, 1775-1789, doi:10.5194/acp-17-1775-2017, 2017.
 - Veefkind, J.P., de Leeuw, G., and Durkee, P.A: Retrieval of aerosol optical depth over land using two-angle view satellite radiometry during TARFOX, Geophys. Res. Letters, 25, 3135-3138, doi: 10.1029/98GL02264, 1998.
- WHO (2018), World Health Organization: http://www.who.int/phe/health_topics/outdoorair/databases/en/, last accessed 16 July 2018.
 - Wang, Y., Yao, L., Wang, L., Liu Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Sci. China Earth Sci., 57, 12-25, doi: 10.1007/s11430-013-4773-4, 2013.
- Xin, J., Wang, Y., Pan. Y., Ji, D., Liu, Z., Wen, T., Wang, Y., Li, X., Sun, Y., Sun J., Wang P., Wang G., Wang X., Cong Z., Song T., Hu B., Wang L., Tang G., Gao W., Guo Y., Miao H., Tian S., and Wang L., The Campaign on Atmospheric Aerosol Research Network of China: CARE-China, Bull. Amer. Meteor. Soc., 96, 1137–1155, doi: 10.1175/BAMS-D-14-00039.1, 2015.
 - Xu, X., Qiu, J., Xia, X., Sun, L., and Min M.: Characteristics of atmospheric aerosol optical depth variation in China during 1993-2012, Atmos. Environ., 119, 82-94, doi: 10.1016/j.atmosenv.2015.08.042, 2015.
 - Xue, Y., He, X., de Leeuw, G., Mei, L., Che, Y., Rippin, W., Guang, J., and Hu, Y.: Long-time series aerosol optical depth retrieval from AVHRR data over land in North China and Central Europe, Remote Sens. Environ., 198, 471-489, doi: 10.1016/j.rse.2017.06.036, 2017.

- Yu, Y., Kalashnikova, O. V., Garay, M. J., Lee, H., and Notaro, M.: Identification and characterization of dust source regions across North Africa and the Middle East using MISR satellite observations, Geophys. Res. Lett., 45, doi: 10.1029/2018GL078324, 2018.
- Zhang, J., Reid, J.S., Alfaro-Contreras, R., and Xian, P.: Has China been exporting less particulate air pollution over the past decade? Geophys. Res. Lett., 44, 2941–2948, doi:10.1002/2017GL072617, 2017.
- Zhao, T. X.-P., Laszlo, I., Guo, W., Heidinger, A., Cao, C., Jelenak, A., Tarpley, D., and Sullivan, J.: Study of long-term trend in aerosol optical thickness observed from operational AVHRR satellite instrument, J. Geophys. Res., 113, D07201, doi:10.1029/2007JD009061, 2008.
- Zhao, B., Jiang, J.H., Gu, Y., Diner, D., Worden, J., Liou, K.-N., Su, H., Xing, J., Garay, M., and Huang, L.: Decadal-scale trends in regional aerosol particle properties and their linkage to emission changes, Environ. Res. Lett., 12, 054021(2017), doi.org/10.1088/1748-9326/aa6cb2, 2017.
 - Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-374, in review, 2018.

Appendix A

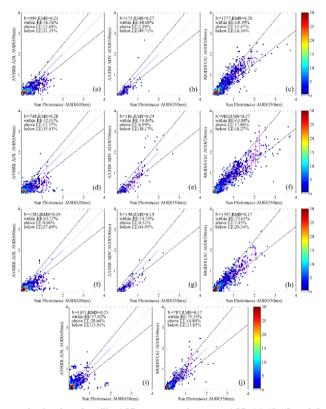


Figure A1: Seasonal scatter density plots of satellite AOD versus sun photometer AOD. (a), (d), (f), and (i) are AVHRR AOD vs. AERONET AOD in Spring, Summer, Autumn, and Winter; (b), (e), and (g) are ATSR AOD vs. AERONET AOD in Spring, Summer, and Autumn; (c), (f), (h), and (j) are MODIS AOD vs. AERONET AOD in Spring, Summer, Autumn, and Winter.

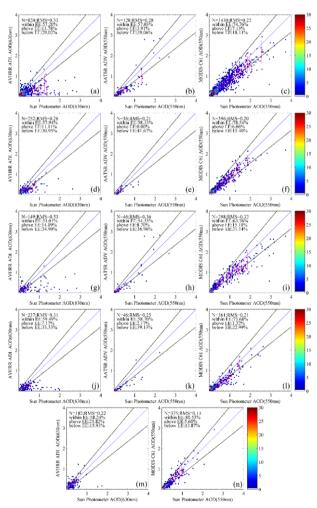


Figure A2: Scatter density plots of satellite-retrieved AOD versus sun photometer retrieved AOD data from Beijing station. ADL all data (a), in Spring (d), in Summer (g), in Autumn (j), and in Winter (m) vs. AERONET, ADV all data (b), in Spring (e), in Summer (h), and in Autumn (k) vs. AERONET, MODIS all data (c), in Spring (f), in Summer (i), in Autumn (l), and in Winter (n) vs. AERONET.

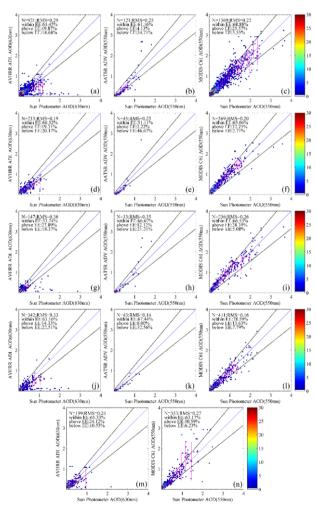


Figure A3: Scatter density plots of satellite-retrieved AOD versus sun photometer retrieved AOD data from XiangHe station. ADL all data (a), in Spring (d), in Summer (g), in Autumn (j), and in Winter (m) vs. AERONET, ADV all data (b), in Spring (e), in Summer (h), and in Autumn (k) vs. AERONET, MODIS all data (c), in Spring (f), in Summer (i), in Autumn (l), and in Winter (n) vs. AERONET.