Interactive comment on "Spatial and seasonal variations of aerosols over China from two decades of multi-satellite observations. Part II: AOD time series for 1995–2017 combined from ATSR ADV and MODIS C6.1 for AOD tendencies estimation" by Larisa Sogacheva et al.

Anonymous Referee #2 Received and published: 11 May 2018

- 10 My main concerns with this paper is the small amount of detail included both on which precisely datasets were used but also on presenting the actual findings. Even though the authors recognise three distinct periods of AOD behaviour in their timeseries, and increase, a plateau and a decrease, they present a number of figures and tables with linear trends on the entire period, which is statistically incorrect. I suggest they re-think this finding accordingly.
- 15 Furthermore, the two datasets, [A]ATSR and MODIS have a clear bias which, although discussed, has not sufficiently been excluded as the reason for the three periods identified. The tendencies calculated appear to be of the same order of magnitude as this bias which leads to the thinking that the bias might be responsible. Consider deseasonalising the datasets before any further analysis is performed.

The constant reference to Part I is also rather tiring and I consider that a few lines of the findings of that Part would go greatly towards improving their comments.

The figures are not of optimal quality, I suggest increasing the pixel quality, and provide ACP with their *eps versions.

The topic is of high interest and merit but I find a number of changes will have to be made on the presentation of the datasets as well as of the findings.

- 25 Refer to the annotated document for further technical, wording as well as scientific comments. Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2018-288/acp-2018-288-RC1supplement.pdf
- 30 Dear Referee#1,

5

Thank you for your criticisms and suggestions to the manuscript. Most of the modifications suggested were considered in the manuscript and below are the comments to your questions/suggestions, which are now marked with **bold**.

P1C1. Please add which AOD this refers to [I assume 550nm?] and then for the rest of the text you can

35 keep it simply as AOD.

- Wavelength is specified

P1C2.

- The preposition is removed

P1C3. This comment applies to all such numbers given in the entire text: I fail to understand how AOD might increase by 2% per annum, i.e. 20% per decade. A change of 0.006 in AOD per annum cannot represent 2% of the total AOD.

Please clarify exactly what you mean and alter accordingly in all locations in the text.

5 Also, it is vital that you add an error estimate in this trend. This estimate is provided easily by all major

languages. Since this paper is based on the trend analysis, a simple error estimate on this trend has to

appear. At least.

- If averaged for the tested period AOD is 0.3, then the annual increase of 0.006 is 2% of the mean AOD. In 10 years, AOD increase is 0.06, which is 20% of mean AOD. That means, that at the beginning of the period, AOD was ~ 0.27, while by the end of the period AOD was 0.33
- 10

P1C4. This seems enormous... please check and alter.

- Please, see my explanation above

P2C1. ca? please use full word.

- Replaces by "approximately"
- 15 P2C2. This is another major issue in my opinion, which has to be addressed centrally and will possibly alter major parts of the rest of the paper. How do you justify a linear trend on a time series that you have shown is made up of a clear positive trend, a plateau and a negative trend? the linear analysis on the entire series has absolutely no meaning. I suggest you rethink the focus of the paper and alter the entire text accordingly.
- 20 Discussion on the whole period anomalies is removed from the manuscript.

P2C3. Re-write.

- See reply to P2C2

P2C4. as expected.

- Added
- 25 P2C5. A further issue I have with this analysis is why did you not deseasonalise the time series before performing the linear trends calculation, as is statistically appropriate. Please comment both here as well as in the text.
 - Deseasonalisation was not needed since yearly averages were used to estimate inter-annual trend and seasonal aggregates for seasonal trends.
- 30 P2C6. Maybe also add information from a more generic review paper, for e.g. https://onlinelibrary.wiley.com/doi/abs/10.1002/9781118682555.ch8
 - The reference is added

P3C1. Three out of four of these are gases, unless you are referring to the particles, in which case, please re-write.

35 - The statement is clarified, references are added.

P3C2. Please provide references that testify that this period was a transitional one.

- The statement is modified.

P4C1. Was this performed with AOD estimates or AI estimates?

- AOD estimates, as mentioned.

P4C2. What did Su et al. find? if you do not add this information, then remove the reference, there is no

- 5 point.
 - The main findings are summarized.

P4C3. What is this acronym? add.

- Added.

P4C4. What did they show? which typical regions? you have to actually explain what the articles you reference are all about, not just mention them.

- The main findings are listed.

P4C5. As above, what did these studies show?

- The main findings are summarized

P4C6. Re-phrase this word. I think "here-after" is more modern.

15 - Re-phrased as suggested.

P5C1. Correct this reference to point to the appropriate ACP[D] paper.

- The reference is corrected.

P5C2. Please expand acronym here, since this is the first time you mention it.

- The acronym is expanded here.

20 P5C3. Either Section or Sect. [see line below] please correct according to ACP wishes.

- Corrected

P5C4. What does this mean? either expand a bit here or do not mention it at all.

- Short clarification added; more in Sect.4

P5C5.

25 - Removed, as suggested

P5C6.

- Removed, as suggested

P5C7. Update all such wordings.

- Updated as suggested.

30 P6C1. Move this explanation above, the first time you mention ADV [not in the title]

- The explanation is moved to Introduction.

P6C2. I fully understand that this is Part II of a two-part paper series, however some information is required so that the reader interested in this paper is not required to have Part I at hand as well. Two sentences should suffice and is not too much work to add.

- As you mentioned, Part 2 is a paper of a two-part paper series. Those papers are closely connected and the authors try to convince the reader to read the Part 1 first. Part 1 is already published in ACP. The main findings from Part 1, which are used in Part 2, are discussed in Part 2 in more detail. The ATSR temporal and spatial coverage is not a main conclusion from Part 1 which is further investigated in Part 2. Thus, we do not think that the more detailed discussion on the coverage is needed in Part 2.
- P6C3. Are you sure this is statistically correct? should you grab all the daily values and average them into a season? in this way, all possible ~90 days of the season will have equal weight to the average. In your case, you are basically weighing the three monthly values, one against the other. Also, how were these monthly means calculated? was there a threshold imposed on the amount of daily values required to compute it?
- 15
- We grab all daily values and average them into a month. Seasonal value is the AOD averaged over 3 months.

With that, each month has the same weight. We assume this is statistically more correct than to average 90

days of data into the season.

P6C3. Are these daily L3 datasets? monthly? please add this quite important information here.

20 - The important information is added.

P7C1. Again, I do not think that one should have to go to a different paper to find this rather important information. The location and choice of AERONET sites is important, as important as which AERONET data were used, level 1, level1b, level2? please add a sentence here on which AERONET sites this refers to.

- Here we just summarize the validation results. The location of the AERONET stations are marked now in Fig.2

P7C2. Please be more precise, what does close in time mean, 5 minutes or 1 h, for e.g.?

- Time is specified.

P7C3. If this main aim of this paragraph was to show the interconsistency of the two satellite datasets, I am sorry to say that you have not reached this goal. Re-phrase this paragraph and re-write adding more

- 30 information, maybe you can refer to Weatherhead et al., 2017, https://doi.org/10.5194/acp-17-15069-201, as well.
 - Part 1 was aimed to show the interconsistency of the two satellite datasets. The main conclusions from Part 1 are summarized in Part 2. We added the reference to Weatherhead et al., 2017

P8C1. Re-write this phrase, does not make sense.

35 - The phrase is re-written

P8C2. Please find a more appropriate title for this section.

- We think that the title fits well to the section, were we discuss the study area and explain the choice for the areas to be compared. We modified the map and moved the section up.

P8C3. Due to what? land cover? land use? please expand.

- The discussion is expanded

P8C4. Re-phrase.

- The sentence is re-phrased.
- 5 P9C1. The purple lines separating the regions do not show. Also, the figure in general is not very clear. I

suggest you increase the resolution.

- The figure is modified.

P9C2. There is a question that arises that is not explained satisfactorily: how can one be sure that these are real trend differences and not inter-sensor algorithm differences that affect the trend? the bias between the two datasets, has it been examined? de-seasonalised? is it flat across the years or does it change? a longer discussion is needed here.

- The bias has been examined in Part I. Deseasonalisation was not needed, as explained in the answer to P2C5.

The section is party re-written for clarification and better understanding.

15 **P10C1.** Main questions on the methodology:

- 1. What do you do when the AOD values are too small, for e.g. 0.0001 and 0.0002, which results in a big, unreasonable, and unphysical corrective factor? do you have a threshold that you apply? explain.
- 20

-

The numbers you mentioned are too low and never considered. AOD accuracy, according to GCOS, should be within ± 0.02 . To avoid too high corrective factor, relative correction is introduced (eg. 4 and eg. 5) where the difference in AOD between ATSR and MODIS is scaled by the AOD.

25 2. Why are you not weighing the AODs with their respective error estimates? or at least, the std?

- We follow Sofieva et al., 2017 who makes the next statement: "different amounts of data available over time result in varying uncertainties over time, which might improperly weight the time series. The reference is added

P11C1. Surely you can spend 5 lines here explaining again. Please recall that Part I is not a published paper, hence one cannot simply reference such a document as if it were the absolute truth. You have to discuss these differences, in short, here.

- Part 1 is published. The sentence is modified by adding the main reasons for disagreement

P12C1. The titles within the graph, DJF, JJA, etc, are hard to read.

35 - Background for the seasonal titles within the graph is modified

P12C2. In this section, you definitely have to discuss the expected seasonal behaviour of AOD over China as well as the relative factors that affect it, are the dominant sources biogenic or anthropogenic, dessert dust, etc.

- This discussion is the main topic of Part I

P12C3. So, you have a clear steady mean bias of 0.1 on annual averages of between 0.4-0.5? i.e. a 20-25% bias?

- 5 I suggest you add a figure of monthly mean values before this figure and help discuss the differences of the two satellite datasets otherwise I am not convinces they can simply be "added as one" observational set.
 - Seasonal maps for ATSR and MODIS AOD, as well as maps for difference between two instruments are shown
- 10 and discussed in Part I. The results from comparison of the validation results is, in our opinion, a prove for the method suggested.

I suggest you take a long look at the reference list of Weatherhead et al., 2017, and draw knowledge from other similar works, for e.g. https://www.atmos-chem-phys.net/14/6983/2014/

15 - Thank you for providing the reference. Even though not referred to that particular manuscript, similar

methods have been used in Part I to discuss the similarity in AOD from ATSR and MODIS

P13C1. You need to provide a reference for this statement and validation results.

- This statement is made in current paper as a conclusion from the results obtained by the authors.

P13C2. Re-phrase.

20 - The paragraph is re-written

P14C1. This is not shown in either Figs 5 and 6. Re-phrase or add Figures/statistics to support your statements.

- Reference to Part 1, supplement, is added

P14C2. This is not a scientific term, re-phrase accordingly.

25 - The whole sentence is deleted.

P15C1. This entire section feels completely out of balance with the rest of the paper and the aims of the rest of the paper. I suggest to the authors to remove it and keep it for another, more policy-oriented article they might write in the future.

30 - The changes in emission policy are only mentioned and not discussed from the political point of view. This

short paragraph is needed, since the goal of the manuscript is to show that changes in the emission policy are

seen in the AOD measurements.

P16C1. Are you using the word tendencies because you are analysing P2 which spans only 6 years and you do not wish to say trend?

- 35
- Exactly. Both periods are shorter than discussed in Weathrehead et al., 1988

P16C2. From this section onwards, I strongly recommend that the authors remove all results/plots/statistics/discussion on the WP. It cannot be accepted as viable statistically.

- The WP results are removed. The reason to show the WP was to compare the results with the other authors, who consider the longer period of 1-2 decades.

P16C3. It is impossible to see the green dots.

5 - The font for green dots was enlarged.

P16C4. This range, -0.1 to 0.1, is the magnitude of the bias between the sensors. How do the authors explain this and how can it be shown that this tendency is a real feature?

- The tendency is calculated for the combined dataset, where the bias between instruments is considered and
- 10 corrected. The correction depends on AOD.

P16C5. How do you show results over the sea when you didn't use data over the sea, as you state in the relevant sections? please check.

15 - The results over sea is removed. The main reason to not discuss AOD over ocean was the luck of the validation points

P22C1. I strongly suggest you move this figure, and the associated discussion, before you discuss Fig. 7.

- In Fig. 7 we introduce the tendencies over whole China pixels-wise (L3) and show where the tendencies are significant. Then we compare AOD tendencies for selected regions. Our logic is to go for details (comparison
- 20 between regions) after introducing the whole picture.

P24C1. This graph should also move further in the front of the paper. It is also rather too busy with all the values/statistics/asterisks etc.

- Please, see our reply to P22C1.
- We agree that the graph is busy but we consider that all information shown is complementary for better overview and the graph is readable. It will take some effort from the reader to read the corresponded numbers in the table.

P26C1.

- The statement is deleted.

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Interactive comment on "Spatial and seasonal variations of aerosols over China from two decades of multi-satellite observations. Part II: AOD time series for 1995–2017 combined from ATSR ADV and MODIS C6.1 for AOD tendencies estimation" by Larisa Sogacheva et al.

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We thank the reviewer Dr. A. M. Sayer for his positive statement and very constructive comments. We much appreciate his thoughts and suggestions that reflected in the improvement of this manuscript. Detailed answers are below.

15

Summary:

I am writing this review under my own name (Andrew Sayer) as I have previously discussed this research with the authors, and am on the team responsible for the MODIS aerosol data products being used in the study. I also reviewed the paper de Leeuw et al (2018), which is in some sense a predecessor to this study, and the Part I of this paper also by Sogacheva et al and also currently in ACPD. I feel I am able to provide an impartial review, but am signing the review in the interests of transparency.

The goal of this pair of papers is to look at spatial and temporal (seasonal/interannual) variations of AOD over China. This is accomplished mainly by using two satellite data sets: the ADV algorithm applied to the combined ATSR2/AATSR record (1995-2012), and the combined Deep Blue/Dark Target algorithms applied to the MODIS Terra record (2000 onwards) from the latest Collection 6.1. Part I contains some validation results and an initial look at the time series, while this Part II focuses on trends ("tendencies" in the authors' terminology) during several time periods where emissions policies may have influenced the aerosol loading. These papers are linked so I will summarize my review of Part I first (which be found on the ACPD page at https://www.atmos-chem-physdiscuss.net/acp-2018-287/), since this Part II requires Part I to stand on. For Part I, I have recommended revisions and re-review. The two main technical threads of my review of Part I were that (1) more needed to be done to establish the validity of treating ATSR2 and AATSR as a single record (which is the underlying but untested assumption), and (2) some of the time

5 series analysis in Part I should be moved to Part II to keep the flow of both papers better and avoid some redundancy. So this review should be read with that in mind.

My overall recommendation for this Part II is also for major revisions and re-review. It's an interesting and important topic, but I don't think it is ready for publication in current form. I would like to review the revision; I would prefer if Part I can be revised and eventually

- 10 accepted for publication first, if possible, so that we have that as a stable version to refer back to when reviewing a revision of this Part II, since the papers are quite closely linked. This is an interesting study but I think (see below) that the ATSR/MODIS merging technique requires some more examination, and also the conclusions would be better supported by including additional meteorological and/or geophysical data products in the analysis (so we
- 15 can see whether AOD changes are likely to be the result of policy, or whether weather patterns may be an influence here).

Uncertainties in the method and results also need better quantification. Note I am not an expert on policy or emissions, so my comments mostly focus on the statistics and AOD data. Hopefully another reviewer can comment on policy/emissions in more detail

20 - my lack of comments is due to a lack of expertise to judge in those areas.

The quality of language is overall good and any issues can probably be dealt with by Copernicus' copy-editing and typesetting process. Therefore my review mainly concentrates on technical abstracts. I have tried to separate each main comment into its own paragraph to respond to. Here, PXLY refers to page X, line Y.

25 **Specific comments:**

Abstract: I would condense this into one paragraph if possible and shorten it to highlight the main findings. For example I think the authors can cut out the discussion on linear trends across the whole period, since one of the main points of the study is that it should not be considered one period due to the changes in emissions policy. I'd also cut out the

- discussion of annual trends/tendencies since I think (as discussed before, and below) only seasonal trends are meaningful due to the seasonal differences in aerosol loading, type, and retrieval coverage (from e.g. cloud cover, snow) aliasing into the annual means in a complicated way. Also, the papers cited in the abstract can be removed these citations are in the body text, they are just adding length here; traditionally one doesn't
- ³⁵ need to provide citations to back up statements in the abstract because that's what the rest of the paper is for.

The Abstract is shortened. The citation is removed.

P3L2-3: I would avoid giving urls like this as citations here, particularly since the latter is an opinion piece. Urls are not always stable and one can't be sure the content is going to change or is valid. It would be better to cite something with a DOI or official publication number. For example the first link is for the World Bank so there must be some report or

something which can be used. 5

URL is replaced with a reference to the similar publication.

Figure 1: Likewise, I would not give an url here for the population data used. If you click 10 through the url, it gives a citation for the data set which should be used instead.

The DOI for the data is provided.

A couple of other things jump out at me from this figure. First, it seems that the largest population change in this region is not in fact China, but India. If population acts as a driver for anthropogenic aerosol emissions, one might expect that observed aerosol

- changes in China may be influenced by changes in transported aerosols from India. If this 15 contribution cannot be guantified, it means that one cannot state that observed changes in China are a result of changes in Chinese policy. (The fact that aerosols don't follow national borders is one reason why in general I prefer regional studies to national studies - you have to be able to account for the broader context of regional
- emissions/meteorology changes.) 20

We agree with the hypothesis that aerosol measured in China might be transported from some other areas (India, Russia, etc.). However, on averaged AOD maps, AOD level over SW China is low, since the AOD transport from India and Bangladesh, which are the strong source of aerosol particles of different origin, is highly blocked in eastern direction by the Himalayas. Whereas highly

- populated and industrial areas are often recognized in China by the local elevated aerosol 25 concentration. The increase in the population usually follows the growth of industry and, in case of China, is resulted in the increase of pollutants. In the current ms, we consider different areas, where the economic growth was not equal during the last decades. Difference between the areas in the aerosol tendencies proves that changes in local emissions play stronger role than the
- change in the aerosol transportation to China from outside the country. We mention later in the 30 text other reasons for changes in the aerosol concentration, but the impacts of each factor are not considered.

Secondly, it looks like the population in the Sichuan Basin area (30 N, 105 E) has dropped somewhat since 2000, while the rest of China has been flat or steadily 35 increasing. Is this right? I did a quick search online and it looks like the Chengdu metropolitan area population is increasing (http://worldpopulationreview.com/worldcities/chengdu-population/ - perhaps this is the red dot on the map here – but the overall population of Sichuan province is fairly stable (http://population.city/china/adm/sichuan/). However I'm not sure of the reliability of these sources, and it is difficult to estimate the total population from these maps of population density because the colour scales seem to saturate and we don't have grid size information. So perhaps people in Sichuan province are becoming more concentrated in Chengdu, I don't know. The point is, I think both of these aspects should be discussed in more detail in the manuscript.

5 You are right that overall the population in the Sichuan province has not changed much. However, people were moving to the Chengdu metropolitan area (current color scale, which was chosen to be the same for all three maps for comparison may not show that clearly) from other regions of the Sichuan province. Thus, we discuss in the text also the changes in population in megacities. We added some clarification to the text.

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Section 2: This is largely a repetition of Part I, in that it is introducing the satellite data used. I understand a recap of the data is needed, but I think that this could be shortened. For example we don't need to know the spectral and spatial resolution of the ATSR and MODIS instruments, or provide the validation summary table. I think it's enough to say

- 15 you're using the level 3 monthly products at 1 degree, and refer back to Part I for more details. Particularly since the number of 3-way matchups (AERONET, ATSR, and MODIS all together) was low and largely confined to cities in Eastern China, I think discussing these statistics in detail gives a (possibly false) impression that we can be confident that these are representative of relative performance across the whole of China. Plus, instantaneous
- 20 validation differences will not necessarily reflect differences in the monthly or longer means, and the method in Section 4 is meant to show and reconcile these differences. So I think Section 2 can be shortened to a couple of paragraphs.

The section is modified. The note about the limited number of validation points in the northwest of China is added. We keep the summary for validation results, on which the method introduced here is based.

is based.

Section 4, general: the large number of long subscripts on variable names makes things cumbersome to read (hard to visually follow the equations). I suggest replacing "ADV" subscripts with "A" and "MODIS" or "MOD" (both are used, but I think mean the same thing) subscripts with "M". Other subscript shortenings could include "y" for "year", "c" for "comb", or "rc" for "rel corr", for example. This will make it easier to follow the

30 for "comb", or "rc" for "rel_corr", for example. This will make it easier to follo equations.

Some of the subscripts are shortened, as suggested. Others are kept as originally introduced since with further shortening the visualization makes reading more complicated.

³⁵ My understanding of the method is that the overlapping ATSR/MODIS period (2000- 2011) just averages the two time series. Then the size of this adjustment in the overlapping period is used to scale the pre-2000 ATSR data, and the post-2011 MODIS data, to generate the "merged" time series. This is done twice: once for an "annual" correction, used for the merged multiannual time series, and once for a set of four "seasonal"

corrections, used for the merged seasonal time series. Further, the calculation is performed separately for each 1 degree grid cell. Is that a correct description? If so I would include a couple of sentences to that effect somewhere up the top, before the equations, in case the reader gets lost.

The text was modified. In the current version, we first introduce the method and then say that it is 5 applied to L3 annual and seasonal aggregates.

P10L27-28: the authors mention Bourassa et al (2014) as an example of this technique being used elsewhere. However l'm not sure that is necessarily a good justification. Bourassa et al were looking at stratospheric ozone, which has (to my knowledge) a somewhat longer lifetime and smoother spatial distribution than AOD, as well as fewer contextual (i.e. surface cover or type-dependent) uncertainties. These are very different error characteristics. Looking through the Bourassa paper, the relative differences between the ozone data sets used were in many cases a lot smaller than the differences seen for AOD here. So the method which is justifiable for one geophysical data set is not necessarily a good choice for another one. Are there other example applications, or justifications which

15 can be made?

We agree that the approach for combining ozone data sets is different (https://www.atmos-chemphys.net/17/12533/2017/acp-17-12533-2017.html). We removed the reference to Bourassa et al. (2014). We could not find any reference where a similar method to combine data from different satellites is introduced and discussed.

20

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I have some more general concerns about this method for combining the ATSR and MODIS records, which become more severe because the purpose is trend analysis, which becomes particularly sensitive to values at the start and end points. Both the start and end points of the combined record are being adjusted by this method, and the "combined"

to "adjusted MODIS" changeover also takes place at the same time (year 2011) as the start 25 of the trend analysis period "P2" (2011-2017). So uncertainties in the merging process will unfortunately affect the trend analysis at the points at which a trend calculation is most susceptible to artefacts.

We indeed did not find the way to estimate the uncertainties related to the merging. However, we estimated the difference in the tendencies, assuming that the combined AOD is over- or 30 underestimated by 10%. In the figure below, we show the annual AOD tendencies for P1 (upper plot) and P2 (lower plot) estimated for each region from the combined dataset (black line) and for two "tested" AOD datasets. In the first tested dataset we assume that our combined dataset is overestimated and "real" AOD is 10% lower (blue line). In the second dataset, we assume that our

combined dataset is overestimated and "real" AOD is 10% higher (red line). For both P1 and P2, 35 the difference in the AOD tendencies for the tested datasets was less than 10% from the tendency estimated of the combined dataset.



However, we decided not to include those checks into the manuscript, since, as it was mention before, the length of the periods, where we estimate tendencies, is short and the results might have uncertainties due to the short length of the periods (thus, called tendencies).

- Instead, to give an estimation of the quality of the combined AOD datasets, in the revised version we added a chapter (Sect. 3.3) on the ADV, MODIS and combined AOD seasonal/yearly comparison with AERONET. This is not validation but more a comparison, since all the AERONET available data were used to calculate the seasonal aggregates and the satellites temporal coverage is lower. It means that some of the events might be missed by the satellites which are included in
- 10 the AERONET AOD seasonal/annual aggregates. The comparison has been performed for four periods:
 - T2 (2000-2011, overlapping)
 - T3 (2011-2017)
 - ADV period (1998-2012)
- 15 MODIS period (2000-2017)

For T1 (1995-2000) there was not enough AERONET data for comparison.

The comparison shows that the statistics for the combined dataset are slightly better than for MODIS and ADV separately.

I appreciate the authors' efforts to merge the ATSR and MODIS records here, and I don't know that there is any well- defined most appropriate way to do so. So this is one attempt which seems like a reasonable thing to try, and I am not sure what other way to harmonise the AOD record to suggest. However the fact remains that this method will introduce uncertainties, which may be systematic, and influence calculated trends. We calculated the AOD standard errors for each year/season for both ADV and MODIS but, we decided to not include it to the manuscript. Here is an example for annual AOD :



For China and SE China, the AOD standard error is low (~0.01). For other areas, the AOD standard error is higher, but what is important here is that the ADV and MODIS AOD standard error time series have similar patterns overall. This gives us the confidence to conclude that the AOD standard errors do not influence the AOD time series significantly.

So, at a minimum, the possible influence of these effects must be quantified. Here are some thoughts about how to do that. The decision to do a simple average for the overlapping (2000-2011) period seems to have been made on the fact that the mean bias of the two data sets vs. AERONET, based on the limited available number of samples and limited available locations of samples, was roughly equal and opposite (so averaging might be expected to cancel out the bias on an average basis). Despite the fact that this was presented as a difference in ABSOLUTE AOD bias, the correction term is applied as a RELATIVE AOD correction. So that feels somewhat inconsistent.

We scaled the correction to avoid the situation, when the correction itself is higher than the AOD.

The authors could make maps of MODIS-ATSR on a monthly basis, both in absolute and relative terms, and see what it looks like. If the bias vs. AERONET of roughly 0.06 (for MODIS) and -0.07 (for ADV) is representative everywhere, then I would expect these maps of absolute difference to have around values of 0.12 and have little angula or temperal.

20 of absolute difference to hover around values of 0.13 and have little spatial or temporal variability. If they don't, then it tells you that the AERONET matchups aren't representative

of the bigger picture. In contrast if maps of relative AOD show small variability, it suggests that a relative scaling is more appropriate.

We disagree that the absolute difference of \sim 0.13 should be representative everywhere. This number depends on the sampling. For fine-dominated aerosols, the validation results show a bias

5 of -0.09 and 0.08 (for ADV and MODIS, respectively); for coarse-dominated aerosols, the bias is -0.11 for ADV and 0.10 for MODIS. The conclusion we draw is that biases for ADV and MODIS are opposite in sign but similar in amplitude. We modified the conclusions in Sect. 3.3.

For the period with both ATSR and MODIS data (2000-2011), my understanding is that the two time series are averaged, and it is the correction across this 11-year period which is

- 10 the basis for correction periods T1 and T3. So one other thing which could be tested is to see what the variability in the MODIS-ATSR difference is over 2000- 2011, e.g. what the standard deviation of the difference is. If it is 0 then MODIS and ATSR are always offset the same amount. In reality it will be nonzero, and this additional variability should be propagated into the trend uncertainties discussed later in the paper. Although you
- 15 might get a trend with apparently low noise, if you know that part of your time series may have uncertainties which aren't captured by this error model, then those errors (in this case, the interannual variability in MODIS-ATSR AOD in 2001-2011) should be added on when estimating the total uncertainty on a trend. A similar point can be made (see my review of Part I) when considering the combination of ATSR2 and AATSR to give the single combined ATSR measurements.
- 20 **ATSR record used as the basis here.**

We estimated the standard error for MODIS-ATSR difference over 2000-2011. See the figure below.



For the annual AOD, the dAOD standard error is around 0.01; for the seasonal AOD, the error is somewhat more significant (ca. 0.02 with some deviation for different seasons and regions). We consider that it is low value and will not change the tendencies significantly (as shown above, where the difference between AOD from the combined data set and the tendencies for the combined $\pm 10\%$ AOD are presented).

In Sofieva et al., 2017 the uncertainties are not considered (https://www.atmos-chemphys.net/17/12533/2017/acp-17-12533-2017.html), where trends in ozone are estimated. They make a statement that "different amounts of data available over time result in varying uncertainties over time, which might improperly weight the time series. In our regression, all data points are considered with equal weights, and the uncertainty of the fitted parameters is estimated from the regression residuals". We are following Sofieva et al. method, and also added uncertainties for the estimated tendencies to the table with the statistics for tendencies.

- 10 My remaining comments are more general, because I think that the above comments and discussion, plus the review of Part I, may necessitate a rewrite of some of the later sections of this study. Some of the points which I think need to be discussed here in more detail include:
- 1. Trend terminology. The authors say "tendency" rather than "trend" throughout. I prefer 15 the more standard "trend". From previous discussions with the authors, my understanding is that they preferred the term "tendency" because they feel that "trend" should refer to a longer time period than considered here. My personal feeling is that "trend" is clearer for the reader, so long as it is made explicit that one should not extrapolate out of the time period under consideration.
- Indeed, we say "tendency" rather than "trend" because the length of the periods when AOD is more less steadily changing is short (11 and 6 years) to estimate trends (Weatherhead et al., 1998).

2. Annual and whole period trends. I continue to think that, since the time series show seasonal variation and are not linear, it is not sensible to do whole period (WP) trends, or annual trends. I think it's best to show only the piecewise trends.

The results and discussion on the WP AOD tendencies were removed. We included those into the version submitted to ACPD thinking that they could have been used for the comparison with other studies (e.g., Wang et al., 2017). However, we keep the results and discussion on the annual AOD tendencies, since this general information is useful for e.g., modelers.

- 30 3. Trend breakpoints. The authors split the trend analysis into WP, an early P1, and late P2. These split times are informed by times where policy changes may have had an influence. However there also exist established statistical methods to estimate whether there are breakpoints in trends, and when these breakpoints are. It would be good to use these methods to see whether in fact any such breakpoints are detectable at the point the
- 35 authors assume they are there.

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In the current manuscript, we study the possible connection between the emission policy and the aerosol concentration. Other factors, e.g., meteorological conditions, dust transport, biomass burning are discussed in Part 1 and Part 2 briefly.

Besides binding the choice for the periods to the Five-Years Plans and emission reduction policy, we performed statistical tests, where we looked at the AOD tendencies, uncertainties of the tendencies, the error of the slopes and tendencies relative error annually and for all seasons all regions. The example for the statistics for annual tendencies for periods of different length starting in 1995 is shown below (number at, e.g., 1999 show statistics for the period 1995-1999).



Similar test was performed for the second breakpoint in ~2011. We added a paragraph to the manuscript (Sect. 5.3), summarizing the results.

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4. Trend significance. The actual fitting mechanism and uncertainty estimation is not discussed in detail. P21L5 says it is linear regression (I assume ordinary least squares) and they define p<0.05 as statistically significant (I assume here this is de- fined as the estimated trend is at least twice as large as the estimated uncertainty in that trend, i.e. 2 sigma). Is autocorrelation considered? Often in trend studies lag 1 autocorrelation is estimated and used to correct the uncertainty estimate (because many geophysical data fields can be autocorrelated on time scales of months, seasons, or years). This gives a more realistic, and generally larger, estimate of the uncertainty on the trend because autocorrelation tends to be positive. See e.g. Weatherhead et al (JGR 1998,

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https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/98JD00995). If this is not done then there should be some evidence given why this model is reasonable.

We have not include autocorrelation into our tendency analysis. The reason for that was that we do not expect any autocorrelation between yearly AOD (as, an example, one exists for other

5 variables dependent on, e.g., 11-years solar cycle). Our annual time series are built from annual values only. Seasonal time series are built from correspondent seasonal aggregates, thus there is a one-year lag between neighboring values.

Further, as noted above, the uncertainty from the ATSR/MODIS merging exercise should ideally be considered.

10 As mention before, we did not have a possibility to estimate the uncertainties. The results for seasonal/interannual comparison with AERONET, which are included in the revised version, gives a rough estimation on the combined AOD quality.

Additionally, there's the problem that since so many comparisons are being performed there are likely to be a number of false positives (trends which appear significant but

15 are coincidental). See e.g. Wilks (JAMC 2006, https://journals.ametsoc.org/doi/10.1175/JAM2404.1) for more on the false discovery rate and how to deal with this sort of thing.

Thank you for the suggestion. In the current manuscript, the AOD *tendencies* were discussed; we apply the statistical significance test for both L3 AOD time series fitting and the averaged

20 over the selected areas AOD. We will consider the method introduced in Wilks when the length of the period will be long enough to discuss *trends*.

5. Presentation of trend uncertainties. In the later Figures and discussion, trends are given in terms of both absolute and relative AOD (where I guess relative AOD is defined with respect to some base year – this is unclear – since as the AOD changes by some absolute amount per year, the relative trend would change while the absolute trend would not). For clarity and comparability between regions, I would rather just see absolute AOD trends.

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We think that the maps with the AOD relative tendencies contain information that is complementary to the AOD absolute tendencies. With those, we can discriminate the areas where the AOD had the higher changes compared with the AOD local values. This information can be potentially compared with relative changes in the emissions (currently we do not have access to that information on the emissions).

I would also like to have the uncertainties estimated on the trends be presented and discussed (the Appendix tables say they have absolute error in percent, but it's not clear

to me what exactly that means, and it is more relatable to have in absolute units like the trend itself). For example at face value the sign of a trend may have changed between two time periods, but it may be that the difference is within the uncertainties of the individual trends, in which case it is more accurate to say that a change in behaviour cannot be identified. Thresholds on p-value are somewhat arbitrary and so I think it is generally more

useful to present trend uncertainties instead (or in addition) in the text and tables in the Appendix.

We added the results for tendencies uncertainties to the Table A1 and discussed them in the text.

- 6. Evidence of attribution for trends. It makes sense that changes in policy could affect emissions and change the AOD. However another key factor, which is examined in several other studies in China and elsewhere, is changes in meteorology. For example if there have been changes in air stagnation frequency, or aerosol transport pathways (for sources outside China as well as those inside China), then these might be magnifying or masking any trends resulting from policy changes. Additional data sets that might be able to support this would include emissions data bases, other satellite products (e.g. SO2 and NOx, which are briefly discussed), and meteorological reanalyses. Some of this has been done by other studies, and I'd like the discussion to go into more detail on those. But some of this may not have been and the authors might need to do these analyses themselves. Otherwise it is premature to try to state the reason for the trends.
- 15

In de Leeuw et al. (2018) and Part 1, we briefly discussed the potential impact of the meteorological conditions and the dust transport on the AOD in China. In the introduction to Part 2, we also shortly mention the results from other studies on the contribution of the meteorological conditions on the AOD in China. E.g., Gu et al. (2018) are referred, who shows that the variation of AOD over Beijing

- 20 was significantly affected by the anthropogenic aerosol emissions and less affected by the wind and temperature inversion. In the current manuscript, we show that the AOD tendencies have a good agreement with the 5-years plan related emission policies. We discuss the changes in the emission policy in China and briefly discuss the changes in NOx and SO2 emissions in China published by van der A (2017). The detailed analysis on the decoupling of the contribution from
- ²⁵ changes in the emissions and the meteorology, including dust transport, and their influence on NOx, SO2, and AOD over the whole China, which implies a detailed analysis of the reanalysis data, is a topic for another manuscript.

In summary, the main thread is I feel it is important to be thorough and give reasonable uncertainties – say what we can – than to make a conclusion which isn't fully supported.

- 30 Particularly since this is an inherently political topic. Maybe the data we have are not enough to be conclusive yet, in which case it is even more important to say as much as we can but no more and be clear about what the biggest uncertainties which we need to reduce to be able to answer the question are. The last sentences of the paper (P26L26-27: "Thus, in the current study the effect of the changes in the emission regulations policy
- ³⁵ in China is evident in AOD decrease after 2011. The effect is more visible in the highly populated and industrialized regions in SE China.") are very strong statements, and this might indeed be the case. But I don't think the discussion of uncertainties is quite thorough enough, or other explanations and their contributions examined in enough detail, to make this case.
- 40 We agree that the statement is strong, but we are not claiming that the AOD decrease is caused

by the changes in emission policy only (we discuss other factors which may influence AOD in the introduction). "Evident", in the current sentence, means that the AOD decrease is noticeable after 2011, when a new regulation plan for emissions was accepted. Those two events (new police and the decrease of the AOD) are in good agreement. The contribution of the anthropogenic emissions

5 to the AOD is critical in China (however, might be comparable with episodic dust or biomass burning transport). Such a strong decrease cannot be caused by e.g., meteorological factors only, which contribute less to the AOD, as compared to the anthropogenic emission in China, as shown by Gu et al. (2018).

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Spatial and seasonal variations of aerosols over China from two decades of multi-satellite observations. Part <u>H2</u>: AOD time series for 1995-2017 combined from ATSR ADV and MODIS C6.1 <u>for and AOD</u> tendencies estimation

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15 Abstract

Understanding long-term trends-variations in aerosol loading is essential for evaluating the health and climate effects of airborne particulates as well as the effectiveness of the pollution control policies. The expected satellite lifetime is about 10 to 15 years. To study the variations of atmospheric constituents over longer periods, the information from different satellites should be utilized.

- Here we introduce a method to construct a combined annual and seasonal AOD-long time series of AOD at 550 nm using the Along-Track Scanning Radiometers (ATSR: ATSR-2 and AATSR) and MODerate resolution Imaging Spectroradiometer Terra (MODIS/Terra), which together cover the period of 1995-2017 period. The long-term (1995-2017) annual and seasonal combined AOD time series are presented for the all of mainland China, for southeastern (SE) China and for 10 selected regions in China and analysed to reveal the AOD tendencies during the last 23 years. Linear regression has been was applied to the combined AOD time series constructed for individual L3 (1°x1°) pixels of the annual and seasonal combined AOD time series to estimate the AOD
- 25 tendencies for three two periods: 1995-2006 (P1) and 2011-2017 (P2), as regarding the changes in the emission reduction policies in China., and the whole period 1995-2017 (WP), when combined AOD time series is available.
 Positive tendencies of annualDuring P1, the annually averaged -AOD-AOD increased with(-0.006 (,-or 2% of the AOD averaged over the corresponding period), per year) prevailed across all of mainland China, before 2006 due toreflecting -emissionincreasing
- emissions increases induced bydue to rapid economic development. In <u>SEsoutheastern</u> China, the annual AOD positive tendency
 in 1995-2006 was 0.014<u>(, or 3% of AOD)</u>, per year in <u>SE China</u>, reaching maxima (0.020, or 4% of AOD, per year) in Shanghai and the Pearl River Delta regions. <u>After 2011, during P2, Negative</u> AOD tendencies reversed across most of China with annually averaged AOD decrease by (-0.015 (, or -6% of AOD), per year) were identified across most of China after 2011 in conjunction
 - with response to effective emission reduction in anthropogenic emissions primary of primary aerosols, SO₂ and NOx (Jin et al., 2016,

van der A et al., 2017). The strongest AOD decrease is observed in <u>the</u> Chengdu (-0.045, or -8% -of AOD, per year) and Zhengzhou (-0.046, or -9% -of AOD, per year) areas, while over the North China plane and coastal areas the AOD decrease was- lower than - 0.03; (or ca-approximately _-6% -of AOD), per year. In the less populated areas, the AOD decrease was small.

- The AOD tendencies for the whole period 1995 2017 were much less pronounced compared to P1 and P2. The reason for that is
 that positive AOD tendency has been observed at the beginning of WP (in P1) and negative AOD tendency has been observed at
 the end of WP (in P2), which partly cancel each other during 1995 2017. In the WP, AOD was slightly increasing over the Beijing-Tianjin Hebei area (0.008, or 1.3% of AOD, per year) and the Pearl River Delta (0.004, or 0.6% of AOD, per year). A slightly negative AOD tendency (0.004, or 0.7% per year) was observed in the Chengdu and Zhengzhou areas.
- Seasonal patterns in the AOD regional long term trend are evident. The contribution of seasonal AOD tendencies in annual tendencies was not equal along the year. While the annual AOD tendency was positive in 1995 2006, the AOD tendencies in winter and spring were slightly negative (ca. 0.002, or 1% of AOD, per year) over the most of China during that period. AOD tendencies were positive in summer (0.008, or 2% of AOD, per year) and autumn (0.006, or 6% of AOD, per year) over all mainland China and SE China (0.020, or 4% of AOD, per year and 0.016, or 4% of AOD, per year in summer and autumn, respectively). The AOD negative tendencies in 2011 2017 were higher compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and and SE China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and compared to other seasons in summer over China (ca. 0.021, or 7% of AOD, per and can be a can be can be a can be a
- 15 year) and over SE China (ca. 0.048, or 9% of AOD, per year). The AOD tendency vary by both season and region. The increase in the annually averaged AOD during P1 was mainly due to increase in summer and autumn in SE China (0.020, or 4%, and 0.016, or 4%, per year, respectively), while during winter and spring the AOD actually decreased over most of China. The AOD negative tendencies in 2011-2017 were larger in summer than in other seasons over China (ca. -0.021, or -7%, per year) and over SE China (ca. -0.048, or -9%, per year).
- 20 The results obtained in the current studylong-term AOD variations presented here show that the effect of the changes in the emission regulations policy in China during 1995-2017 is evidentresult in <u>a AOD</u>-gradual decrease of the AOD after 2011 with an average reduction of 30%-50% between 2011 and 2017. The effect is more visible in the highly populated and industrialized regions in SE China, as expected.

1 Introduction

- 25 Atmospheric aerosols play an important role in climate change through direct and indirect processes. In order to evaluate the effects of aerosols on climate, it is necessary to study their spatial and temporal distributions. Understanding the long-term changes and the trend in AOD on the Earth, especially in the developing countries like China, becomes increasingly essential for accurately assessmenting the radiative forcing, as well as better constraining the climate models effects on climate (Li et al., 2013). The rapid development of industry, traffic and urbanization, the combustion of fossil fuel, the emissions of industrial fumes and contaminated
- gas lead to a significant increase of atmospheric aerosols, which do not only affect climate, but also constitute a threat to human health (<u>Tie et al., 2009;</u> Cao et al., 2017). It is critical for environmental and epidemiological studies to accurately investigate the

fine-scale spatial and temporal changes in aerosol concentrations regarding the industrialization and urbanization (Streets et al., 2009; Kanakidou, 2014).

Satellite aerosol remote sensing is a rapidly developing technology that may provide good temporal sampling and superior spatial coverage to study aerosols. The most common parameter derived from satellite observations is the Aerosol Optical Depth (AOD),

5 which is a measure of the extinction of electromagnetic radiation at a given wavelength due to the presence of aerosols integrated over the atmospheric column. AOD is a key factor for the estimation of the aerosol concentration, the evaluation of atmospheric conditions and the effect of atmospheric aerosols on climate.

The air pollution in China is severe (Bouarar et al., 2017), widely distributed, and the atmospheric chemical reactions <u>are</u> complex (Kulmala et al., 2015). The strong economic growth in China (<u>http://www.worldbank.org/en/country/china/overviewWorld Bank</u>,

10 <u>2017</u>; Morrison, 2018) has significantly raised the living standards (<u>http://www.china.org.cn/opinion/2013-10/24/content_30391004.htmZhang K., 2017</u>), but it has also brought serious environmental damage and degradation (Tang et al., 2015).

In 1970–1990, the dominant contributing sources were big and small coal burning stoves widely used in power plants, industry, utilities and households (Jin et al., 2016). Coal smoke mainly contains sulphuric acid (SO₂), total suspended particle matter (TSP),

- but also nitrogen oxides (NO_x) and carbon monoxide (CO), which affect air quality by atmospheric aerosol formation (Siihto et al., 2006; Fuzzy et al., 2015; Sarrafzadeh et al., 2016; Nzihou and Stanmore, 2015). Other sources, such as dust from construction sites, are mainly consisting of primary PMs, contributed less. During the transition period from 1990 to –2000, the growing number of vehicles, mainly in megacities contributed a lot to the increase in NO_x and volatile organic compounds (VOCs). From 2000 to the present, the anthropogenic air pollution is ingrained in the megacities and spreading to the regional level (Jin et al., 2016). However,
- 20 national <u>and</u> regional plans, environmental laws, rules and standards exist in China, which are revised with the Five-Years-Plans, which are series of social and economic development initiatives (Jin et al., 2016).

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The further urbanization is a consequence of the <u>a</u> growing industry. Over the periodDuring <u>-1978the 1978</u>-2016 period, more than 550 million migrants moved to China's cities, resulting in a large rise of urban population from 18 to 57% (Zhang, 2017). <u>CThe</u> coastal regions where manufacturing and services are better developed, especially big cities, <u>wereare</u> the <u>major principal</u> destinations (Chan, 2012). During the last two decades, a strong population inflow to eastern China has been reported (Ma and Chen, 2012;

- Center for International Earth Science Information Network CIESIN Columbia University, 2017). The population in the North China Plane, the Yangtze River Delta and the Pearl River Delta was steadily increasing mainly due to the growth of large cities, or metropolises (Fig. 1) on account of migration from the less industrialised areas (see also Kourtidis et al., 2015; Stathopoulos et al., 2017). The population density has increased in the Beijing-Tianjin-Hebei area, Shanghai, Xiamen and Guangzhou, by more than
- 30 200% <u>in during</u> 2000-2015 while in Wuhan, Chengdu and Zhengzhou the population has grown <u>by</u> nearly by 50%. Such a strong population growth has resulted in the fast urbanization and the further industrialisation and infrastructure development in those regions.







-Figure 1. The population density in China (doi: 10.7927/H4DZ068D) in 2000 (a) and the population density change in 2005 (b), 2010 (c), 2015 (d) compareding with 2000. Megacities are marked with green circles. -Data are obtained from https://doi.org/10.7927/H4DZ068D, accessed 09.02.2018.

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Due to the sparse surface network in China and limited data availability, the satellite remote sensing is increasing used to study the long-term changes in aerosol properties over China. Satellite remote sensing is a rapidly developing technology that may provide a good temporal sampling and a superior spatial coverage to study aerosols. The most common parameter derived from satellite observations is the Aerosol Optical Depth (AOD), which is a measure of the extinction of solar radiation at a given wavelength due to the presence of aerosols integrated over the atmospheric column. AOD is a key factor for the estimation of the aerosol

- 10 to the presence of aerosols integrated over the atmospheric column. AOD is a key factor for the estimation of the aerosol concentration, the evaluation of atmospheric conditions and the effect of atmospheric aerosols on climate. Due to the sparse surface network in China and limited data availability, the increasing attention has been paid over the last decades to the satellite aerosol remote sensing to study the long time changes in aerosol properties over China. Su et al. (2010) analyzed the AOD distribution over 10 locations in East Asia using the yearly mean AOD products from POLDER (Polarization and the effect of atmospheric sensing to study the long time changes in aerosol properties over China.
- 15 Directionality of the Earth's Reflectance) radiometer during the period from 2005 to 2009 and showed that the spatial distribution of fine-mode aerosols over East Asia is highly associated with human activities. Xie and Xia (20092008) reported statistically increasing AOD trends in spring and summer in north China annually during the period from 1982 to 2001 using the Total Ozone Mapping Spectrometer (TOMS). They also demonstrated the increasing tendency of AOD (500 nm) from 1980 to 1991 and a reverse tendency from 1997 to 2001 in north China.Su et al. (2010) analyzed the AOD distribution over 10 locations in East Asia using the
- 20 yearly mean POLDER_AOD products during the period from 2005 to 2009. Guo et al. (2012), who analyzed the monthly the AOD trends of TOMS and MODIS in China during the period from 1982 to 2006 in 1982-2008, have reported similar findings. The spatial variation in AOD suggests no apparent upward trends in 1980's; since 1990, both TOMS and MODIS indicate a significant AOD increase across China (Guo et al., 2012). The temporal and spatial trends in AODs were analyzed with MODIS data (2000-2008) over eight typical regions in China (Guo et al., 2011). Seasonal patterns in the AOD regional long-term trend are revealed. As shown
- 25 by Guo et al. (2011), AODs exhibit mostly similar seasonality during the period from 1980 to 2001: AOD maxima is observed in winter (except for Taklimakan Desert), in summer the AOD is higher than in autumn; while there is not such seasonality in 2000– 2008. Li et al. (2014)Opposite in sign AOD trends-reported opposite AOD trends in different locations in China have been recorded using observations from the Aerosol Robotic Network (AERONET, Holben et al., 1998) by Li et al. (2014). Many authors reported decreasing trends in the United States and Europe, and iIncreasing trends in China over the last decades in the last few years, as
- 30 summarized in Zhao are revealed -by Wang et al. (2017), while a few recent studies (Zhang et al., 2016; He et al., 2016; Mehta et al., 2016; Zhao et al., 2017; Zhang J. et al., 2017) showed decreasing trends in China in the last few years over the last decade. The expected satellite lifetime of the satellites is about 10 to -15 years. To study the longer trends of the substances using satellites, the information from different-variations of atmospheric constituents over longer periods, the information from different satellites

should be <u>combinedutilized (Weatherhad et al., 2017)</u>. Combining multiple sensors could increase the period for data availability and reduce data uncertainties (Li et al., 2016).

In this study, we introduce <u>the a</u> method to combine <u>seasonal and annual AOD data retrieved from</u> the Along-Track Scanning Radiometers (ATSR-2 and AATSR, <u>hereunder here-after</u> referred as ATSR) and <u>the MODerate</u> resolution Imaging

- 5 Spectroradiometer Terra (MODIS/Terra) to create multi-decadal time series coveringseasonal and annual aerosol data for_the period from 1995-to 2017 to investigate AOD tendencies in China-during more than two decades. The method is based on the results from the comparison between ATSR and MODIS AOD products presented in (Sogacheva et al. (,-2018), further referred as Part IPart 1). We also investigate whether the tendencies in AOD are related to emission changes as well as to the pollution control policies in China. Since In view of the spatial AOD variations across difference exists in aerosol loading in China (Part I, de Leeuw et al.,
- 10 2018; Part 1, and references cited there in), we present-the combined AOD-long time series and estimate AOD tendencies for different regions in China.

The objectives of this study are: (1) to combine <u>AATSR Dual View (ADV, Ver2.31)</u><u>AATSR ADV</u> and MODIS/<u>Terra collection</u> <u>6.1 (C6.1)</u> seasonal and annual aerosol data during 1995–2017 and (2) to analyze the spatial and temporal <u>variations of the seasonally</u> and annually <u>averaged AOD aerosol long time series</u> and link the AOD tendencies to emission control policies in China.

15 The paper is structured as follows. The AATSR and MODIS/Terra AOD products are <u>briefly</u> introduced in Sect. ion 2, including the short description of the instruments, aerosol retrieval algorithms, <u>data setdataset</u>s and validation results.- (for more details see Part 1). The study area is discussed in Sect. 3, where 10 selected regions are introduced. In Sect. 4, a method <u>used is presented</u> to construct the combined from ATSR (1995-2011) and MODIS (2000-2017) <u>seasonal and annual AOD time series for more than 20 years</u> (1995 2017) is presented, and the AOD correction applied to ATSR and MODIS AOD is discussed (Sect. 4.1), the, long-term annual

20 and seasonal combined AOD time series are shownare introduced (Sect. 4.2), --and evaluated with the AERONET. (Sect. 4.3). In Sect. 5 we present and discuss the results for the AOD evolution over China (Sect. 5.1), the and relate them to changes in the emission reduction policies in China are discussed(Sect. 5.2). T; the AOD tendencies are estimated for three periods related to the changes in the emission policies in China for selected periods and regions are presented and discussed in Sect. 5.3 and discussed. The main conclusions are summarised in Sect. -6.

25 <u>2 Study area and selection of the regions</u>

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China covers a huge territory with significant regional differences (Fig. 2). Climate in China varies from subarctic (north) to subtropical (south). The west and north of the country are dominated by deserts (such as the Gobi and the Taklamakan), rolling plateaus, and towering massifs. The southern areas of the country is hilly and mountainous terrain. The eastern plains and southern coasts of the country consist of fertile lowlands and foothills. The contradictions between the economic growth and the environmental quality have varying dimensions in different regions. The largest agricultural provinces are Henan, Sichuan, Hunan, Anhui, and Jiangsu with low population growth and Shandong, Heilongjiang, and Hubei with balanced urbanization and agricultural

development. Corresponding differences among regions exists by means of land urbanization (Lin et al., 2015). The highest

population density is observed in the east and southeast (Fig. 1). Cities with rapid land urbanization are mainly distributed in the coastal regions and also scattered throughout the inland regions. Sparsely populated areas are provinces in the western China. Therefore, from a regional perspective, there are large differences in the levels of economic development in China and the efficiency of the SE China is far higher than that of the Central and the Western regions. Additionally, the gap between them tends to expand

5 (Yang and Wang, 2013).

AOD over the oceans or islands is not included.

As in Part 1, in current study we focus on the entire area of mainland China, i.e., the area between $18-54^{\circ}$ N and $73-135^{\circ}$ E defined as $1^{\circ} \times 1^{\circ}$ grid cells with retrievals over land and constrained by the borders indicated by the black line in Fig. 2. Purple lines indicate SE China, defined in this study as the over-land area between $20^{\circ} - 41^{\circ}$ N and $103^{\circ} - 135^{\circ}$ E. The numbers indicate the ten study regions. Regions 1-7 nearly cover the SE China. Region 8 covers the Taklamakan Desert, region 9 is over the Tibetan Plateau, and region 10 is over the northeast (NE) China. Note that all areas used in this study only consider the AOD over mainland China, i.e.

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50°N 40°N 30°N 20°N $75^{\circ}E$ 90°E
105°E
120°E
120°E
135°E

15 Figure 2. Regions over mainland China (indicated with the purple numbers and red borders) selected for study of seasonal, interannual and long-term behavior of the AOD and the locations of the Aerosol Robotic Network (AERONET) sites (yellow diamonds) used in this study for validation. The black line indicates mainland China. A larger area over southeastern China is indicated by SE and purple border. With some deviations, the choice of the regions is similar to those in other studies (e.g., Luo et al., 2014, Wang et al., 2017). This choice covers major urban/industrial regions such as the Beijing-Tianjin-Hebei (BTH) area, the Yangtze River Delta (YRD) and the Pearl River Delta (PRD), Sichuan/Chongqing as well as cleaner regions in the northwest (region 10 in Fig. 2) and southeast (region 3). The Tibetan Plateau and the Taklamakan Desert regions, which are an important source of dust particles in China, were chosen to represent the sparsely populated and less developed, in terms of industrialization, regions.

23. AOD data: instrumentation, AOD retrieval algorithms and aerosol data setdatasets

Below we briefly introduce <u>the ATSR</u> and MODIS instruments, AOD retrieval algorithms and <u>aerosol-the AOD data setdatasets</u>. For more details <u>(e.g., difference in ATSR and MODIS AOD spatial and temporal coverage)</u>, see de Leeuw et al. (2018) and <u>Part IPart 1</u>.

23.1 ATSR ADV

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The ATSR instruments, ATSR-2 on board the European Remote Sensing satellite ERS-2 (1995-2003) and AATSR on board the environmental satellite Envisat (2002-2012), <u>hereafter together</u> referred <u>hereunder</u> as ATSR, were developed to provide <u>the</u> high-accuracy measurements of the Sea Surface Temperature (SST), and However, they are also successfully used for atmospheric aerosol retrievals (e.g., Flowerdew and Haigh, 1996; Veefkind et al., 1998; Sayer et al., 2010; de Leeuw et al., 2015; Kolmonen et al., 2016; Popp et al., 2016). Together, these instruments provided 17 years of global data. Both satellites flew in a sun-synchronous descending orbit with a daytime equator crossing time of at 10:30 LT (ERS-2) and 10:00 LT-local time (ENVISATEnvisat). The ATSR is a dual view instrument. One view is near-nadir and the other one is at a 55° forward angle. -The time between the two views is approx. 150 seconds along the track. The nominal resolution at nadir is 1x1 km² and the swath width is 512 km, which results in global coverage in 5-6 days. ATSR has three wave bands in the visible – near infrared (centred near 555 nm, 659 nm, 865

- nm) and four bands in the mid- to thermal infrared (centred near 1600 nm, 3700 nm, 10850 nm, 12000 nm). Over land, the AATSR <u>Dual View (ADV) ADV</u> AOD retrieval algorithm uses the two ATSR views simultaneously to eliminate the contribution of land surface reflectance to the TOA radiation to and retain the path radiance in cloud-free scenes (Veefkind et al., 1998, Kolmonen et al., 2016, Sogacheva et al., 2017) following Flowerdew and Haigh (1996).
- ATSR-2 AOD data are available for the period from June 1995 to- December 2003, with some gaps in 1995 (from January to May, globally) and 1996 (from January to June, globally), while also toward the end (approx. from autumn 2002) the data are less reliable. AATSR data are available for the period May 2002 April 2012, but some data are missing in 2002 and therefore we use AATSR data only from August 2002 onforward. The consistency between the ATSR-2 and AATSR data set<u>datasets has beenwas</u> discussed in Popp et al. (2016). Over China, the difference between the ADV AOD values retrieved from ATSR-2 and AATSR is small, as
- 30 shown by pixel-by-pixel and monthly aggregate comparisons as well as validation results (Part 1). This allows for the combination

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of the ATSR-2 and AATSR AOD time series into one dataset without offset correction over China.- The ATSR temporal and spatial coverage, as regarding seasonal and annual AOD aggregates over China, is discussed in details in Part I.

The L3 (averaged on a grid of $1^{\circ}x1^{\circ}$) seasonal aggregates were obtained for winter (DJF), spring (MAM), summer (JJA) and autumn (SON) by averaging the L3 monthly aggregates to corresponding seasons. The annual AOD data were obtained by averaging the monthly AOD data. Hereunder, the ATSR ADV Ver. 2.31 AOD product will be referred to as ADV.

2<u>3</u>.2 MODIS

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The MODIS/Terra-(MODerate resolution Imaging Spectroradiometer) sensor (Salomonson et al., 1989) aboard NASA's Terra satellite <u>ishas been</u> flying in a near-polar sun-synchronous circular orbit for more than fifteen years since December 1999. MODIS/Terra has a daytime equator crossing time at 10:30 <u>LT-local time</u> (descending orbit), a viewing swath of 2330 km (cross track) and provides near-global coverage on a daily basis. Its detectors measure 36 spectral bands between 0.405 and 14.385 µm,

- 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 (Kaufman et al., 1997; Remer et al., 2005; Levy et al., 2013) and one for retrieval over water surfaces (Tanré et al., 1997; Remer et al., 2005, Levy et al., 2013). The DB algorithm (Hsu et al., 2004, 2013) was traditionally used
- 15 over bright surfaces where DT performance is limited (e.g. deserts, arid and semi-arid areas) and was further developed for returning aerosol measurements over all land types (Sayer et al., 2014).
 - In this paper, MODIS/Terra AOD C6.1 DTDB merged (L3) data (2000 2017, https://ladsweb.modaps.eosdis.nasa.gov/) are used, which are slightly different from C6 over snow and sea ice, where the cloud mask has been improved (https://earthdata.nasa.gov/earth observation data/near real time/download nrt data/modis nrt#ed collection 61).
- 20 improvement is important over northern and central elevated China, which is periodically or all year around covered by snow. Hereafter, the MODIS/Terra AOD C6.1 DTDB merged AOD product will be referred to as MODIS.

23.3 ADV and MODIS validation results.

and it acquires data at three spatial resolutions (250m, 500m, and 1000m).

AOD Level 2 (L2, 0.1° x 0.1° resolution) validation with the AERONET -L2.0 (quality-assured) AOD results over China for ADV (de Leeuw et al., 2018) and MODIS C6.1 (Part I) is discussed in detail in Part 1. The location of the AERONET stations used in this

- 25 study for validation is shown in Fig.2. Note that the number of the AERONET stations is limited over north-west of China (de Leeuw et al., 2018; Part 1). Validation -are results are briefly summarized below in this section. To evaluate the quality of the ADV and MODIS AOD, these products were compared with reference ground based AOD data
- available from AERONET sites (Holben et al., 1998) in the study area (see Table 1 in de Leeuw et al. (2018) for AERONET locations used for validation). For this comparison, collocated data, i.e. satellite data within a circle with a radius of 0.125° around
 30 the AERONET site, were averaged and compared with averaged AERONET data measured within ± 1 hour of the satellite overpass time (Virtanen et al., 2018). The ADV and MODIS AOD validation results are summarised in Table 1.

In this validation exercise, For all available collocations-_between ADV and AERONET and between MODIS and AERONET <u>AERONETwere considered (" (Table 1, "</u>All points"). The results show that the MODIS algorithm performs slightly better than ADV____The correlation coefficients are 0.88 for ADV and 0.92 for MODIS (note, that since MODIS has better coverage as compared to ADV (Part 1), the number of validation points for MODIS is considerably larger). Both algorithms show similar (in absolute numbers) bias, which is negative for ADV (-0.06) and positive for MODIS (0.07). AOD standard deviation (δ) and root_-mean_square_error (rms) are slightly lower for MODIS. Note, that since MODIS has better coverage as compared to ADV (Part 1), the number of validation points for MODIS. Note, that since MODIS has better coverage as compared to ADV (Part 1), the number of validation points for MODIS is considerably larger (4963 and 1132 for MODIS and ADV, respectively). Similar in absolute values but different in sign AOD bias was obtained also for "fine-dominated" (AOD>0.2, AE>1) and for "coarsedominated" (AOD>0.2, AE<1) aerosol conditions (-0.11 and 0.10, for ADV and MODIS, respectively).

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Table 1. ADV and MODIS AOD validation results over China for the overlapping period 2000-2011. Statistics (number of points (N), correlation coefficient (R), bias, standard deviation (σ) and root_-mean_-square <u>error</u> (rms)) for all validation points, <u>separately for "fine-dominated" (AOD>0.2, AE>1) and "coarse-dominated" (AOD>0.2, AE<1) aerosols</u>, and collocated points (*when ADV and MODIS overpasses are <u>close in timewithin ±90min from each other</u> and ADV, MODIS and AERONET retrieve AOD) are shown. For collocated points, statistics are aggregated also seasonally for winter (DJF), spring (MAM), summer (JJA) and autumn (SON).

			_							
	N		R		bias		σ		rms	
	ADV	MOD	ADV	MOD	ADV	MOD	ADV	MOD	ADV	MOD
All points										
	1132	4963	0.88	0.92	-0.07	0.06	0.007	0.003	0.24	0.20
<u>e.g. fine-</u> dominated	482	1983	0.85	0.89	-0.09	0.08	0.014	0.005	0.30	0.24
e.g., coarse- dominated	129	970	0.85	0.88	-0.11	0.10	0.032	0.007	0.37	0.22
Collocated* Points:										
All collocated points	255		0.92	0.93	-0.11	0.06	0.01	0.008	0.17	0.16
Collocated points, seasons:										
DJF	10		0.92	0.96	-0.04	-0.17	0.023	0.052	0.10	0.19
MAM	87		0.81	0.81	0.00	0.13	0.012	0.013	0.16	0.14
JJA	73		0.94	0.96	-0.13	0.13	0.029	0.017	0.25	0.22
SON	85		0.92	0.88	-0.02	0.05	0.007	0.009	0.10	0.09

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To compare the performance of <u>the</u> two algorithms when both ADV and MODIS retrieve AOD, we carried out the AOD validation for the cases, <u>where when the</u> difference between ATSR and MODIS/Terra overpasses was less than 90 minutes and both ADV and MODIS have successfully retrieved AOD around AERONET (<u>Table 1</u>, "Ceollocated points"). Altogether, 255 collocations exist between ADV, MODIS and AERONET for the ATSR and MODIS/Terra overlapping period (2000-2011) over China. Validation

was done for all collocated points and respectively for each of the four seasons: DJF, MAM, JJA, and SON. For all collocated points, the correlation coefficients (R) were similar for ADV and MODIS (0.92 and 0.93, respectively), ADV was biased negatively (-0.11), while MODIS was biased positively (0.06). In winter, MODIS showed a strong negative bias in AOD (-0.17), while the correlation for MODIS is was higher than for ADV (0.96 and 0.92, respectively). Note, that the number of collocated points is was

- 5 low (10) in winter. In spring, R was the same (0.81) for MODIS and ADV, while bias is 0 for ADV and 0.013 for MODIS. Interestingly, both MODIS and ADV showed similar tendency of underestimation of AERONET AOD for AOD >0.6 in spring. -In summer, R was a bit higher for MODIS; biases-was equal for ADV and MODIS were equal in an absolute numbers-sense but opposite in sign (negative for ADV and positive for MODIS). In autumn, R was a higher for ADV (0.92-against versus 0.88 for MODIS), the bias was negative for ADV (-0.02) and positive for MODIS (0.05).
- 10 To conclude<u>In summary</u>, MODIS show<u>eds a better performance for the</u> "all points" selection. Similar (in absolute numbers) bias, which is negative for ADV-(0.07) and positive for MODIS_a-(0.06) will beis considered in Sect. 4, where <u>a</u> method for combining the two datasets will beis presented. For collocated points, the better performance of either ADV or and MODIS is not clearly pronounced showed similar performance.

3. Study area and selection of the regions

- 15 China covers a huge territory with significant regional differences (Wang et al., 2017), with corresponding differences among regions by means of land urbanization (Lin et al., 2015). corresponding differences among regions by means of land urbanization (Lin et al., 2015). Corresponding differences among regions by means of land urbanization (Lin et al., 2015). The contradictions between economic growth and environmental quality have varying dimensions in different regions. More urbanized areas have faster rates of growth and are probably the major contributors to air pollution. The highest population density is observed in the northeast, followed by the east (Fig.1). Cities with rapid land urbanization are mainly distributed in the coastal regions and scattered throughout the inland regions. The largest agricultural output provinces are Henan,
- Sichuan, Hunan, Anhui, and Jiangsu with low population growth and Shandong, Heilongjiang, and Hubei with balanced urbanization and agricultural development. Sparsely populated areas are provinces in the western China. Thus, from a regional perspective, there are large differences in the levels of economic development in China and the efficiency of the Eastern region is far higher than that of the Central and the Western regions. Additionally, the gap between them tends to expand (Yang and Wang, 25 2013).
- Thus, to study the spatio temporal tendency of AOD in China during the last more than 20 years and to examine if the differences in economic activity among the regions are reflected in AOD level and tendencies, we focused on the entire area of mainland China, SE China and ten typical areas, as shown on Fig. 2 (adapted from Part I). The mainland China is the area within the Chinese border indicated by the blue line. SE China, defined in this study as the over land area between 20°-41° N and 103°-135° E, is indicated
- 30 by purple lines. Numbers indicate the ten study regions. Regions 1–7 nearly cover the SE China. Region 8 covers the Taklamakan desert, region 9 is over the Tibetan Plateau, and region 10 is over the NE China. Note that all areas used in this study only consider the AOD over mainland China, i.e. AOD over the oceans or islands is not included.



Figure 2. Regions over mainland China selected for further study of seasonal, interannual and long term behaviour of the AOD, overlaid on the ATSR-retrieved (ADV version 2.31) 12-year aggregated AOD map. Mainland China is indicated with the blue line. The figure shows 10 regions over China and a larger area over SE China indicated with SE (as in Part I).

With some deviations, the choice of the regions is similar to those in other studies (e.g., Luo et al., 2014, Wang et al., 2017). This choice covers major urban/industrial regions such as the Beijing Tianjin Hebei (BTH), the Yangtze River Delta (YRD) and the Pearl River Delta (PRD), Sichuan/Chongqing as well as cleaner regions in the north (region 10 in Fig. 2) and southeast (region 3). In addition, the Tibetan Plateau and Taklamakan desert regions were chosen to represent the sparsely populated and less developed, in terms of industrialization.

4- Long-term (1995-2017) annual and seasonal AOD time series combined from ADV and MODIS

Here we introduce a method to combine the AOD data from the ATSR (1995-2011) and MODIS/Terra (2000-2017) radiometers, which together cover the period from 1995 to -2017. The results from the comparison of the ADV and MODIS AOD datasets comparison in Part IPart 1 are used here to construct a combined AOD dataset. Below, some conclusions obtained in Part IPart 1 are bighlighted

- 15 are highlighted.
 - (1) Similar AOD patterns are observed by ADV and MODIS in yearly and seasonal aggregates (Part IPart 1). However, the ADV-MODIS difference maps (Part IPart 1, Fig. 7 (right) and Fig. 10 (right)) show that MODIS AOD is generallymostly higher than that from ADV.
 - (2) The time series in Figs. 9-13 and S1-S4 (Part IPart 1) show largeshow large differences between regions, for both sensors, while the interannual patterns in the time series are similar for both ADV and MODIS.

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- (3) Similar patterns exist in year-to-year ADV and MODIS annual AOD tendencies in the overlapping period (Part IPart 1, Tabs. 2 and S2).
- (4) ADV and MODIS validation with AERONET <u>data (Part IPart 1</u>, Sect.<u>3.3.4</u>) shows similar high correlation (0.88 and 0.92, respectively), while the bias is of similar magnitude but opposite in sign: positive for MODIS (0.06) and negative for ADV
- (-0.07). Similar in absolute values but opposite in sign AOD bias is calculated for "fine-dominated" (-0.09 and 0.08, for ADV and MODIS, respectively) and for "coarse-dominated" aerosol conditions (-0.11 and 0.10, for ADV and MODIS, respectively).

4.1 Method

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The combined AOD (AOD_{comb}) time series database AOD_{comb}, 1995-2017- for the period from 1995 to 2017 have been was compiled

from AOD estimated for three periods.:+tThe first period (T1) is the pre-EOS period (1995-1999), when only ATSR was available, the second period (T2) is the ATSR and MODIS/Terra overlapping period (2000-2011) and the third (T3) is the post-ENVISAT (2012-2017) period, when only MODIS/Terra was available:

$$AOD_{comb,1995-2017} = [AOD_{T1}, AOD_{T2}, AOD_{T3}].$$
 (1)

First, we introduce the combined AOD for the overlapping period T2-(AOD_{T2,vear}), when AOD for both ADV (AOD_{ADV}) and

15 MODIS (AOD_{MOD}) is was available. AOD for each year ($AOD_{T2,y,year}$) is calculated as a mean of AOD_{ADV,y} and AOD_{MOD,y} AOD of ADV and MODIS:

$$AOD_{T2,y;year} = \frac{AOD_{MOD,y;year} + AOD_{ADV,y;year}}{2} . (2)$$

The simple averaging has been was applied, since ADV and MODIS show similar biases of different opposite signs (Sect. 2.3 and Part 1).

Using ADV and MODIS yearly AOD from the T2, the AOD correction (AOD_{corr}) is was calculated as the mean difference between ADV and MODIS for the overlapping period:

$$AOD_{corr} = \frac{\sum_{years} \frac{AOD_{MOD,y_{y},year} - AOD_{ADV,y,year}}{2}}{N_{years}} \quad ,- \quad (3)$$

25 where N is the length of the overlapping period in years.

For ADV and MODIS respectively, the AOD correction has been was scaled by the corresponding AOD, averaged over the overlapping period:

$$AOD_{rel_corr,ADV} = \frac{AOD_{corr}}{mean(AOD_{ADV,T22000-2011})}, \quad (4)$$
$$AOD_{rel_corr,MODIS} = \frac{AOD_{corr}}{mean(AOD_{MODIS,T22000-2011})}. \quad (5)$$

For T1 and T3, the AOD relative correction (AOD_{rel corr}) has been was applied as positive correction for usually lower ADV AOD (eq. 6) and negative correction for usually higher MODIS AOD (eq. 7)

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$$AOD_{T1,y,year} = AOD_{ADV,y,year}^{*}(1 + AOD_{rel_corr,ADV}), (6)$$
$$AOD_{T3,y,year} = AOD_{MOD,y,year}^{*}(1 - AOD_{rel_corr,MODIS}) (7).$$

*(1 + 100

- AOD

A similar type of merging of data from two separate instruments has been used e.g., Bourassa et al. (2014) to create a long ozone time series.

The method introduced above has been applied pixel wise to annual and seasonal AOD aggregates from ADV and MODIS to construct the combined AOD time series. For each season, has been computed separately, as introduced for yearly AOD.

4.2 AOD relative correction

The spatial distributions of the relative correction for MODIS AOD (AOD_{rel_corr,MODIS}) is are shown in Fig. 3 (for annual AOD aggregates) and in Fig.4 (for and seasonal AOD aggregates). As expected, the highest AOD relative correction (30-40%) corresponds to the areas, where the agreement is lower between ADV and MODIS AOD (Fig.s. 7 and 10 in Part IPart 1), e.g., over

- the bright surface areas, such as Tibetan Plateau and the Taklamakan and Gobi Deserts and Harbin area. The reasons for 15 disagreement between ADV AOD and MODIS AOD, related mostly to the coverage and validation results, is discussed in detail in Part I Part 1 and Sect. 3.3 of the current paper. For seasonal aggregates, the highest correction (ca 45%) is obtained over the Tibetan Plateau in autumn, when AOD is lower compared to spring and summer (Fig. 10 in Part 1). In summer, the correction is smaller, since in that season the agreement between the in ADV and MODIS AOD is better compared tothan in other seasons (Fig.s. 7 and
- 20 10 in Part 1). Over SE China, the AOD correction is lower (between 10% and 20% of AOD). The ADV AOD relative correction shows similar spatial patterns and thus therefore is not discussed here.





Figure 3. AOD relative correction for MODIS <u>seasonal (DJF, MAM, JJA, SON) and annual (Year)</u> AOD for the combined AOD dataset obtained with the method introduced in Sect.4.1

5 Similar to annual, the seasonal AOD correction spatial patterns are recognized. In summer, the correction is smaller, since in that season the agreement in ADV and MODIS AOD is better compared to other seasons (Figs. 7 and 10 in Part I). For seasonal aggregates, the highest correction (ca 45%) is obtained over the Tibetan Plateau in autumn, when AOD is lower compared to spring and summer (Fig. 10 in Part I).



Figure 4. AOD relative correction for MODIS seasonal AOD for the combined AOD dataset obtained with the method introduced in Sect.4.1

The method was applied pixel-wise to L3 annual and seasonal AOD aggregates from ADV and MODIS to build the yearly and seasonal database of combined AOD. For each season, the AOD relative correction was computed separately, as introduced above for annual AOD.

4.3.2 Long-term (1995-2017) yearly and seasonal AOD time series combined from ADV and MODISResults

In this section, we introduce the ADV, MODIS and resulted combined annual and seasonal AOD time series for China and SE China. The long term time series of the seasonally and annually averaged AOD from ADV (red circles), MODIS (green circles) and combined AOD (yellow rhombs)_from ADV and MODIS are shown for the mainlandall China of China and SE part of all mainland China in Fig.5. Since ADV was negatively biased and MODIS is was positively biased with respect to the AERONET AOD, and the biases are similar in absolute value, -the combined time series in pre-EOS and post-Envisat periods are practically corrected by increasing ADV and lowering MODIS AOD with the AOD correction as introduced in Sect. 4.1. As expected, the similar

15 interannual variations in the separate datasets are reproduced in the combined time series.

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Figure 5. ADV (red circles), MODIS (green circles) and combined from ADV and MODIS (yellow rhombs) annual AOD time series for China (left) and SE China (right). The method used to combine the ADV and MODIS time series in presented in Sect. 4.1.

5 An increase in AOD is observed in China from 1995 towards 2006, with a relative peak in 2003, when active forest fires, which occurred over Russia, strongly increased the AOD in NE China. Between 2006 and 2011, the AOD was not showing a clear tendency. After 2011, the AOD started to steadily decrease, with the exception for year 2014. In SE China, the AOD tendencies are more pronounced and show similar temporal behavior.

In Fig. 6, the long term time series of the averaged over the seasons (winter, DJF; spring, MAM; summer, JJA; autumn, SON) AOD

10 from ADV (red circles), MODIS (green circles) and combined (yellow rhombs) from ADV and MODIS are shown for all mainland China and SE part of the mainland China.

-For all seasons, except spring, the difference between ADV and MODIS AOD was low, thus is small and -therefore, the combined time series closely reproduce-resembles both the ADV and MODIS AOD. In spring, the combined time series show, on average, 0.1 show on average lower AOD as compared to MODIS (which, in relative numbers, is about 10-20% lower than the MODIS

15 AOD). However, as for annual aggregates, <u>the interannual variability</u> for seasonal combined AOD the interannual variability in the time series is similar for both ADV and MODIS.

The annual and seasonal AOD tendencies for the combined time series are estimated in Sect. <u>-65</u>.

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Figure <u>4</u>. ADV (red circles), MODIS (green circles) and combined from ADV and MODIS (yellow rhombs) seasonal AOD time series for China (left) and SE China for winter (DJF), spring (MMA), summer (JJA) and autumn (SON)<u>and the whole year</u>. The method used to combine the ADV and MODIS time series <u>in is</u> presented in Sect. 4.1.

4.3 Comparison with AERONET

Combined AOD datasets were built from L3 data for seasonal and annual aggregates, thus the comparison with the AERONET is possible for the corresponding periods. We call the current exercise as comparison rather than validation, since the temporal coverage might be different in seasonal and annual aggregates for AERONET, ADV, MODIS, and combined AOD, which might

- 5 bias the aggregated value. The combined AOD was constructed from three periods (T1-T3, see section 4.1 for details), where different corrections were applied, thus we perform the AOD comparison for the corresponding periods and also for the full ATSR and full MODIS periods. For T1, the number of AERONET locations is limited to perform the comparison (Part 1). For T2, the agreement between AERONET and combined AOD seasonal and annual aggregates (Fig.5, Table A1) is slightly better than between AERONET and ADV or MODIS separately. R is higher in DJF, JJA and year, 1σ and rms are lower for all seasons
- 10 and year. For T3 (Table A1), R is considerably higher in spring (0.49 and 0.61 for MODIS and combined, respectively). Bias, slope, 1σ and rms are lower for all seasons for combined AOD compared to MODIS AOD. For 1998-2012, which covers the period when both AERONET and ADV are available, correlation R is higher for combined AOD in winter, autumn and year; rms is lower in all seasons and year for the combined AOD.

For 2000-2017, which covers the period when both AERONET and MODIS are available, comparison results are similar as in T3.



Figure 5. Density scatterplot of ADV AOD (left), MODIS AOD (middle) and combined AOD (right) versus AOD from AERONET stations over China for the period T2 (2000-2011). The filled circles are the averaged AOD binned in 0.1 AERONET AOD intervals (0.25 for AERONET AOD>1.0) and the vertical lines on each circle represent the 1-σ standard deviation. Statistics in the upper left corner indicate correlation coefficient R, bias, standard deviation, root mean- square error (rms) and the number of data points (N). The colorbar on the right indicates the number of data points in each bin.

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5. AOD tendencies during for the 1995-2017 period.

5.1 Evolution of AOD over China: annual anomaly maps

To study the temporal evolution of the AOD over China during the whole period (WP) from 1995 to 2017, the anomaly maps (Fig.

- 5 6) were created by subtracting the multi-year aggregate for the WP from the yearly aggregates. The evolution of the AOD can be followed for different regions and regions with similar or different evolution of the AOD can thus be identified.
 Strong negative anomalies over SE China (and the north of India and Bangladesh, which are out of the scope of the current paper) in 1995-2001 show that during that period the AOD was lower compared to the AOD averaged over the WP. Towards 2006, the AOD anomalies were becoming less negative: the yearly AOD was increasing and the anomaly decreased. Starting from 2006, a
- 10 positive AOD anomaly, which show that the yearly AOD was higher than the AOD averaged over 2000-2011, was observed over SE China. Between 2006 and 2011, the positive anomalies were staying high. Starting from 2011, the anomaly changed from positive to negative, i.e. the AOD decreased. Starting from 2015 the anomaly was mostly negative getting stronger towards 2017, which was the last year in our studies.

The AOD anomaly pattern in SE China was very variable. In the BTH area (region1), the anomaly was strongly negative in 1995,

15 which was the starting years in the time series, and gradually decreased towards 2006, when it turned to positive. In the area over the YRD (region 2) encompassing large urban and industrial developments such as Shanghai and Nanjing, the multi-year average appeared to be quite representative for the AOD with negative anomalies until 2000, after that the anomalies fluctuated around zero with neither very high nor very low values.

Similar AOD behavior was observed in the Sichuan/Chongqing area (region 6) with a strong positive anomaly in 1997 and 1999,

20 followed by negative anomalies in 2000-2002. After 2002, no significant deviations were observed until 2005 when the AOD exceeded the multi-year average and this situation remains, with small fluctuations until 2014 when a strong negative anomaly expanded over the wider region of the SE China during the next (2015-2017) years. A "belt" of positive AOD anomaly was observed east from the Taklamakan Desert in 2001, which was likely the year of the most

intensive dust event during the period of interest. The widespread positive AOD anomaly in 1998 and 2003 in the northeast of China

25 <u>was likely the consequence of the forest fires over Russia.</u>



Figure 6. AOD anomaly maps for the years 1995-2017 calculated for combined AOD. Anomalies were calculated for each year by subtracting the multiyear (1995-2017) average from the annual aggregates.

- 5 Although meteorological conditions play an important role in mixing and transporting pollutants, increasing local aerosol emission from human activities is generally considered as the dominant cause of increasing wintertime haze over China in past decades (Yin et al., 2017). One can isolate meteorological changes from emissions changes by normalizing the aerosol concentrations by aerosol concentration efficiencies (Yang et al., 2018). However, the contribution from wind and its associated languishing patterns explain the historical increase of regional AOD by 10%, while other meteorological contributions show no significant trends over 35 years
- 10 (Gu et al., 2018).

In the Beijing area, the variation of the AOD is significantly affected by anthropogenic aerosol emissions (Gu et al., 2018) during the last few decades. As seen from the combined AOD time series in Figs. 5 and 6, the annually and seasonally averaged AOD in China was increasing from the first year of the AOD available in our analysis (1995) until 2006 (with some delay for different regions). Between 2006 and 2011, AOD was slightly fluctuating. After 2011, the AOD started to decrease until the end of the study

5 period in 2017. In the current paper, we aim To find out ifto show that changes in the AOD follow the emission control policy in China. tendencies relate to the emissions regulation in China during that period, Below we present a short overview of the emission regulation policies in China during the last two decades.

5.1-2 Emissions regulation policies in China during the last two decades.

Jin et al. (2016) showed that in China: (1) the early policies, until 2005, were ineffective at reducing emissions; (2) during 2006– 2012, new instruments, which interact with political incentives, were introduced in the 11th Five-Year Plan. However, emission control policies on air pollution have not been strongly reconsidered in the 11th Five-Year Plan, thus no significant reduction of the air pollution has been observed. Regional air pollution problems dominated by fine particulate matter (PM_{2.5}) and ground level ozone (O₃) emerged. Nevertheless, the national goal of reducing total sulphur dioxide (SO₂) emissions by 10% was achieved (Lin et al., 2010).

- -Jin et al. (2016) also showed that SO₂ emissions, as well as smoke and dust emissions, have been gradually decreasing since 2006. However, the reduction of the total emission of SO₂, a single primary pollutant, does not necessarily improve air quality. NOx emissions continued to grow which is explained by the growing number of vehicles mainly in megacities. Total NOx emissions in East China reached their peak levels in 2012, and have stopped increasing since then (van der A et al., 2017), when filtering systems were installed, mainly at power plants but also for heavy industry. These regulations for NOx were announced in 2013 in the Air Pollution Prevention and Control Action Plan (CAAC, 2013).
- The "total control" of SO₂ and NO_X is strengthened and accelerated in China (Zhang et al., 2017). As an example, the change in standards for cars in the period-2011–2015 reduces the maximum allowed amount of NOx emissions for on-road vehicles by 50 % (Wu et al., 2017). Further emission control scenarios exist in China to control the entirely coal-burning thermal power plants exists. The newly designed control policies considered in Wang et al. (2018) are predicted to lead to reductions in January levels in Beijing
- 25 between -8.6% and -14.8% for PM_{2.5}, PM₁₀, NO₂, and SO₂. However, regional differences in emission control exist. More strict regulations for on-road vehicles (e.g. a ban on older polluting cars) were introduced on a city level, e.g. in Beijing, rather than nationwide (van der A et al., 2017).

In June 2013, the State Council issued the Action Plan for Air Pollution Prevention and Control. This document laid out the roadmap for air pollution prevention and control across China for 2013-2017. In 2016, the second report was published

30 (http://cleanairasia.org/wp-content/uploads/2016/08/China-Air-2016-Report-Full.pdf) showing the considerable improvements in emission reduction and air quality. The air quality mostly improved <u>at-in</u> the developed regions. This report finds that the cities that failed to attain the 2015 air quality target-and show slow progress and still suffer from poor air quality are concentrated in Henan Province, Shandong Province, and in the Nnortheast of China. Compared with the more developed regions, tThese cities had less experience and insufficient capacity in air pollution prevention and control <u>than the more developed regions such as the BTH</u>-area. Such regional differences might result in some deviation of regional emission tendencies compared to those averaged over the whole China.

Thus Therefore, three periods, closely related to the Five-Year-Plans (before 2006, 2006-2011, and after 2011), can be identified, when the emission reduction policies in China are reconsidered.

5.2-3 AOD seasonal and annual tendencies for the selected periods: 1995-2006 and -2011-2017 and 1995-2017.

In addition to selecting the study periods based on emission reduction policy according to the Five-Years Plans, we performed statistical tests, where we looked at the AOD tendencies, uncertainties and errors on annual/seasonal bases for different periods for

10 all selected regions. The results (not shown here) prove that, with some exception (1-2 years shift for a few regions, depending on the season) years 2006 and 2011 can be chosen as a pivot points for the AOD tendencies in China. Linear regression has beenwas applied to individual L3 pixels of the combined AOD time series to estimate the AOD tendencies over China for three two periods: 1995-2006 (P1) and 2011-2017 (P2), as regarding the changes in the emission reduction policies,

.and the whole period 1995 2017 (WP), when the combined AOD time series is available. For P1 and P2, Tthe AOD tendencies

- 15 (dAOD) averaged over the yearsper year have beenwere estimated, since P2 is too short to estimate the decadal tendencies. <u>Results</u> for 2006-2011 are not shown here since AOD tendency was close to 0. The statistical significance for the tendencies has beenwas estimated with the t_-test (Chandler and Scott, 2011). The results were considered significant at p-value<0.05. We also estimated relative tendencies, which are the ratio of tendencies to the corresponded and the corresponding time series averages (Schönwiese and Rapp, 1997).</p>
- 20 5.2.1 Annual AOD tendencies over China.

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The AOD tendencies and AOD relative tendencies for annual aggregates for P1,—and P2 and WP–are shown in Figs._7 and 8, respectively. AOD positive tendencies are <u>coloured withdenoted by</u> red; AOD negative tendencies are <u>coloured withdenoted by</u> blue. The pixels for which the tendency is statistically significant are marked in Fig. 7 with the green dots.—when the linear fit was giving the statistically significant results (p-value<0.05).

- 25 In P1 (Figs. 7 and 8, left), the AOD increase in summer (JJA) and autumn (SON) with 0.1 to 0.5 (ca. 3-7% of the AOD averaged over the corresponding period) per year contributed most to the annual AOD increase over SE China. In spring (MAM), the AOD tendency was positive over the Yangtze River Delta, northern China and the west of the Tibetan Plateau, (between 0.1 and 0.3, or 2-4%, per year) and negative over the other parts of China. In winter (DJF), when the coverage of the satellite data is lower compared to other seasons (Part 1), irregular patterns of both positive and negative AOD tendencies are observed over China.
- 30 For annual aggregates, a statistically significant increase in the AOD of 0.02 to 0.04 (3-6%) per year (Fig. 8, left panel), was observed in P1 over SE China. A similar increase of the AOD was observed over the Taklamakan Desert. However, since the annual AOD over the Taklamakan Desert is lower compared to the SE China (Fig. 10 in Part 1), the relative tendency over the Taklamakan Desert is higher (up to 10% per year for certain pixels) than over SE China for annual AOD. Note that for only few pixels over the

Taklamakan Desert the linear fit provides statistically significant results. Over western China, the AOD tendencies were slightly negative (between 0 and -0.01 per year). Over northern China they were slightly positive (between 0 and 0.4 per year) for most pixels. However, the results are not statistically significant since p value is larger than 0.05 almost everywhere in those regions. In P2 (Figs. 7 and 8, right), the AOD tendencies were opposite in sign, compared to P1. The strongest AOD decrease (up to -0.16)

- 5 per year) was observed over the northern part of SE China in summer. AOD decrease was slightly lower for other seasons over SE China, while for central and western China the AOD tendencies were close to zero in P2. The relative AOD tendency was between -5% and -10% per year in winter and spring over SE China and more than -15% per year over the Sichuan region in summer and autumn. For annual AOD aggregates, a statistically significant decrease in AOD of -0.02 to -0.04 per year (Fig. 7, right), with an average reduction of 30%-50% between 2011 and 2017, was observed in P2 over SE China. In the Sichuan and Henan regions the
- 10 AOD decrease more with -0.1 per year. The most negative AOD tendency (ca. -15% per year) is observed in northern China over the east of Inner Mongolia area (Fig. 8, right). A small AOD decrease is observed over the Taklamakan Desert; a small AOD increase was observed over the most NE part of China. Over other areas, the AOD tendencies in P2 were close to zero. The low significance of the tendencies can be explained by the low coverage (mostly in winter and spring over snow-covered areas, Part 1), short length of the periods considered and large interannual variations. To reveal more detailed differences in AOD
- 15 tendencies over China, we apply similar fitting to annual and seasonal AOD averaged over the selected regions in China. The results are presented and discussed in Sect. 5.4.





Figure 7. AOD tendencies (per year, see colorbar) from <u>seasonally and</u> annually aggregated combined AOD time series for <u>three_two</u> periods: 1995-2006 (P1) <u>and</u> -2011-2017 (P2). <u>and 1995-2017 (WP)</u>. Individual pixels are marked with <u>the a</u> green dots when the <u>linear fit</u> was giving the tendency is statistically significant according to the student t test results (p_-value<0.05).





Figure 8. AOD relative tendencies (per year, see colorbar) from <u>seasonally and</u> annually aggregated combined AOD time series for three two periods: 1995-2006 (P1) and, 2011-2017 (P2). and 1995-2017 (WP).

A statistically significant increase in AOD of 0.02 to 0.04 per year (Fig.7, left panel), which is about 3–6% of AOD per year (Fig.8, left panel), was observed in P1 over SE China. Similar increase of AOD was recognized over the Taklamakan Desert. However, since the annual AOD over the Taklamakan Desert is lower compared to the SE China (Fig. 7 in Part I), the relative tendency over the Taklamakan Desert is higher (up to 10% per year for certain pixels) than over the SE China for annual AOD. Note that for only

5 few pixels over the Taklamakan Desert the linear fit provides the statistically significant results. Over western and northern China, the AOD tendencies were slightly negative (between 0 and -0.01 per year) over western China and positive (between 0 and 0.4 per year) for most of the pixels over northern China and p value was above 0.05 almost everywhere in those regions.

In P2 (Figs.7 and 8, middle panel), the AOD tendencies were opposite in sign, compared to P1. A statistically significant decrease in AOD of 0.02 to 0.04 per year (Fig.7, middle panel), or between 7% and 15% of AOD per year (Fig.8, middle panel) was
 observed in P2 over SE China. In Sichuan and Henan regions the AOD decrease was higher, reaching 0.1 per year. The highest negative AOD tendency (ca. 15% of AOD per year) was observed in northern China over the east of Inner Mongolia area. Small

AOD decrease was observed over the Taklamakan Desert; small AOD increase was observed over the most NE part of China. Over other areas, the AOD tendencies in P2 were close to zero.

The opposite in sign and similar in absolute values AOD tendencies in P1 and P2 partly cancel each other, when interannual AOD

15 tendencies for the whole period are considered. Over most of the China, the AOD tendencies estimated for 1995 2017 are close to zero. Slightly positive AOD tendencies (ca. 0.1, or 2% of AOD, per year) are observed in the BTH, Shandong and Henan regions.

5.2.2 Seasonal AOD tendencies over China.

Long term AOD variations and their effects on the AOD seasonality over China have been discussed in Part I. Here we show the AOD seasonal tendencies, which are not always similar to the interannual tendencies.

- 20 In P1 (Figs. 9 and 10, left panel), the AOD increase of 0.1 0.5 (ca. 3 7% of AOD) per year in summer (JJA) and autumn (SON) contributed most to the annual AOD increase in P1 over SE China. In spring (MAM), the AOD tendency was positive over Yangzi River Delta (0.1 0.3, or 2 4% of AOD, per year), northern China and west of the Tibetan Plateau and negative over other parts of China. In winter (DJF), when the coverage of the satellite data is lower compared to other seasons (Part I), irregular patterns of both positive and negative AOD tendencies are observed over China.
- 25 In P2, the AOD decrease is observed in all seasons over the most part of China (Figs. 9 and 10, middle panel). The strongest AOD decrease (up to -0.16 per year) was observed over the northern part of SE China in summer. AOD decrease was a bit lower for other seasons over SE China, while for central and western China the AOD tendencies were close to zero in P2. The relative AOD tendency was between -5% and -10% per year in winter and spring over SE China and more than 15% per year over the Sichuan region in summer and autumn.
- 30 Similar to the annual AOD tendencies, the opposite AOD seasonal tendencies in P1 and P2 are partly cancelled by each other in WP (Figs. 9 and 10, right panel). The low AOD decrease (between -0.01 and -0.03 per year) has been observed in WP in spring in the Sichuan region. A small AOD increase (0.01 0.03 per year) was observed over BTH in winter and summer and over Henan in autumn. For the other parts of China, the AOD tendencies for the WP are close to zero. Small negative relative AOD tendencies

(between 3% and 5% per year) were observed in Inner Mongolia in spring. Similar positive relative AOD tendencies (between 3% and 5% per year) were observed in the NE of China. For other seasons and areas, the AOD relative tendencies were close to zero. However, the linear fitting of the individual L3 pixels was not often providing statistically significant results (as indicated by lack of green dots for corresponding pixels) and the single pixel peaks in AOD tendencies are not considered here. The low significance

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of the tendencies can be explained by the low coverage (mostly in winter and spring over snow covered areas, Part I), short length of the periods considered and high interannual variations. To reveal more detailed differences in AOD tendencies over China, we apply similar fitting to annual and seasonal AOD averaged over the selected regions of China.



Figure9. AOD tendencies (per year, see colorbar) from seasonally aggregated combined AOD time series for three periods: 1995-2006 (P1), 2011-2017 (P2) and 1995-2017 (WP). Individual pixels are marked with the green dots when the linear fit was giving the statistically significant results.



Figure 10. AOD relative tendencies (per year, see colorbar) from seasonally aggregated combined AOD time series for three periods: 1995-2006 (P1), 2011-2017 (P2) and 1995-2017 (WP).

5.34 AOD tendencies for the selected regions

5.34.1 Annual AOD tendencies

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In Figure: <u>11-9</u> we show the annual AOD combined time series (black line) for China, SE China and the 10 selected regions (corresponding red numbers in the left upper corner for each region), which are defined in Sect. <u>32</u>. The linear fitting has been

5 applied to AOD for two periods, P1 and P2 (dashed lines). For P1 and P2, we also show the AOD tendencies per year and the corresponding relative AOD tendencies. For P1 and P2, we show the AOD tendency (dAOD) per year. The AOD tendencies and the metrics describing the results for linear fitting for P1 and, P2 and WP are summarised in Appendix, Table A1A2. For both periods, the results for linear fitting were of high confidence tendencies are statistically significant(p value <0.05), except</p>

For both periods, the results for linear fitting were of high confidence tendencies are statistically significant (p-value < 0.05), except for regions 8-10, where, as discussed in Part IPart 1 (Table S1), ADV has low coverage and MODIS has difficulties in retrieving AOD over bright surfaces.

- As expected, the AOD tendencies (dAOD) and AOD relative tendencies were positive in P1 in all chosen regions, except for the sparsely populated Tibetan Plateau (region 8), where AOD is very low and undergoes varies very little from year_to_year variations. The maximum AOD increase in P1 (0.020, or 4% of AOD, per year) was observed in the Shanghai area (region 2) and the PRD and Guangxi province (region 7). In the BTH (region 1), and Hunan and -Guizhou (region 4), dAOD was also high (0.016, or 3% of a start of the start
- AOD, per year). Those regions strongly contributed to the AOD increase in SE China (0.014, or 3%-of AOD, per year). In regions 1, 2, 4, 7 and for all China, uncertainties of the AOD tendencies were within 25% of the AOD tendency in P1. For regions 3, 5, 6 and SE China, the uncertainties related to the tendency estimation were between 25% and 50% of the AOD tendency. In regions 8-10, the uncertainties were higher, which can be explained by the low AOD and lower AOD coverage.
- In P2, the AOD decrease has been<u>was</u> observed in all selected regions (1–10) and thus <u>also</u> in SE China and all of China. In absolute numbers, dAOD was almost twice higher in P2 than in P1. The most rapid AOD decrease (ca. -0.045, or -8%-of AOD, per year) was observed in central regions of SE China (regions 4 and 6), while for the rest of SE China, including regions 1,_2,_3,_5 and 7, dAOD was about -0.03, or ca -6%-of AOD, per year. For regions 8-10, which are less populated and less industrialized, the dAOD was lower (-0.002, -0.014 and -0.004, respectively, or -2%, -5% and -1% per year). <u>Uncertainties related to the AOD tendency</u> <u>estimation were within 25% of the AOD tendency in regions 1 to 7, SE China and China. In regions 8 and 10, the uncertainties were</u> above 50%.



Figure 11.9. Annual AOD long-tTime series (black line) of the annually averaged AOD combined from ATSR and MODIS for MODIS for China, SE China and 10 selected regions (see Fig. 2, correspondingent red numbers in the left upper corner for each region). Results for linear fitting of AOD are shown for two periods, P1 (1995-2011) and P2 (2011-2017), marked with black arrows in upper left subplot. Fitting lines (dashed lines) and AOD tendencies (numbers) are shown in red, when AOD tendency was positive and in blue, when AOD tendency was negative; * shows whether the tendency is statistically significant. The correspondinged relative AOD tendencies for each fit are shown in brackets (in black, %).

5.34.2 Seasonal AOD tendencies

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10 As shown in Part IPart 1 and Sect 5.3.2, spatial and temporal patterns in the seasonal AOD differ from that of the annual AOD. To examine if the AOD tendencies in different seasons equally contributed to variations of the the annual AOD year to year changes or

if the AOD year-to-year changes-variations are more pronounced in certain seasons, we applied similar regression analysis for the seasonal aggregates of AOD for P1 and P2. As for the annually averaged AOD (Fig. <u>119</u>), Fig. <u>12-10</u> shows the seasonally combined AOD time series for China, SE China and the 10 selected regions. We also show the linear fitting of AOD for two periods, P1 and P2. For P1 and P2, we show the AOD tendency-(dAOD) per year and the corresponding relative AOD tendencies. Time series, linear fitting line and the AOD tendency-aretendency are shown in colours related to different colours for each season.

- 5 linear fitting line and the AOD tendency aretendency are shown in colours related to different colours for each season. In P1, during winter and spring, AOD does not have large temporal variability vary much (within ±0.01 per year) in winter and spring in all-any of the regions in China (Fig.1210). <u>AIn winter,</u> negative AOD tendency of -0.017, or -3%, of AOD per year was observed in the Sichuan area (region 6) and in regions 9 and 10, where AOD coverage is low in winter and high AOD variability is observed. In the Hunan and Guizhou Provinces and PRD areas (regions 4 and 7) <u>Small negative AOD tendency</u> the AOD decreases
- 10 <u>a little (by ca -0.006 (, or -2% of AOD)</u>, per year<u>.) is observed in the Hunan Guizhou and PRD areas (regions 4 and 7). In other regions it increases somewhat and small positive (ca 1-2% per year) in other regions. In spring, dAOD was slightly positive in regions 4, 8 and 10 and slightly negative in other regions. In summer and autumn, The the AOD increase was discovered in all regions (1 7) in eastern China (regions 1 to 7) in summer and autumn. In regions 1, 2, 5 and 6 (north and west of the SE China region, respectively) the AOD positive tendency was increased stronger in summer (ca 0.020, or 3-4% of AOD, per year). In regions</u>
- 15 3, 4 and 7 (central east to southern part of SE China region) dAOD was stronger-larger in autumn, increasing towards south and reaching maxima (0.032, or 6%-of AOD, per year) in region 7. -Over the whole of SE China, dAOD was 0.020, or 4%-of AOD, per year in summer and 0.016, or 4%-of AOD, per year in autumn, while in winter and spring dAOD was close to zero-from year to year. In western and northern China (regions 8-10), year-to-year changes in the seasonal AOD in P1 were low (within ±0.01 per year). Over allthe whole China, AOD showed an increase of 0.008, or 2%-of AOD, and 0.006, or 3%-of AOD, per year in summer
- and autumn, respectively, and <u>low-a small</u> decrease (ca. -0.02, or -1%-of AOD, per year) in winter and spring. <u>ThusTherefore</u>, summer and spring contribute most to the annual AOD increase (Fig. 47) during <u>the</u> years 1995-2006 (P1) and AOD changes were considerably higher in SE China <u>than in compared to</u> other areas-of <u>China</u>. Note, that the p-value for linear fit applied here was often above larger than 0.05 (Appendix, Table <u>A2A3) and the results were thus not statistically significant</u>. However, similar AOD tendencies in neighboring regions (with similar population density and economic activity and meteorological conditions) allow

25 making the overall conclusions.

In SE China, which includes regions 1-7, the <u>a</u> strong AOD increase (0.020, or 4% of AOD, per year) was observed in summer. The second maxima was revealed<u>occurs</u> in autumn (0.016, or 4% of AOD, per year). In winter, AOD increase was small (0.001 per year), while in spring a small AOD decrease (-0.003, or -1% of AOD, per year) has been was observed.

AOD tendencies averaged seasonally over all of China for P1 show similar AOD increase in summer (0.008, or 2%-of AOD, per 30 year) and in-autumn (0.006, or 3%-of AOD, per year). In winter and spring, AOD decreased was-slightly decreasing (by - 1%-of

AOD, per year). AOD tendencies uncertainties were smaller than 50% of the AOD tendencies in regions 2-7, SE China and China in JJA and SON. In other seasons and regions, the uncertainties were >50% of the AOD tendencies.



Figure <u>1210</u>. AOD seasonal long-time series (solid lines), linear fitting (dashed lines) and AOD tendency (numbers) for P1 (1995-2011) and P2 (2011-2017) are shown in <u>different</u> colours related tofor each season (blue for DJF, winter; purple for MAM, spring; green for JJA, summer; and light blue for SOA, autumn); * shows whether the tendency is statistically significant. The correspondinged relative AOD tendencies for each fit are shown in brackets (in black, %).

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In P2, the AOD tendency-tendencies over selected regions were was negative and about twice stronger (in absolute numbers) than in P1-over the selected regions. AOD tendencies uncertainties were lower in P2 than in P1. However in regions 8-10, the AOD tendencies uncertainties were >50% of the AOD tendencies in all seasons. In winter, the AOD decrease was high (between -0.024 and -0.029 per year) in regions 1, 2, 5. The relative AOD tendency was of 11% per year in region 5, which is west of the BTH area, between -4% and -6% per year over SE China and regions 1-7, and lower over other areas. In spring, the AOD has been decreaseding stronger compared tothan in winter in regions 3, 4, 6 and 9. The A high negative relative tendency (from -6% to -9%) has been was observed in regions 4-9 in spring. -In summer, the a high negative AOD relative tendency (between -10% and -12%) was observed in the southern part of SE China, regions 4-7. The highest absolute AOD decrease (-0.082, or -9% of AOD, per year)

- 5 was observed in summer in the BTH area (region 1). In autumn, the AOD tendencies were lower compared tothan in spring and summer in most of the regions. In SE China, which includes regions 1-7, the strongest AOD decrease (-0.048, or -9% -of AOD, per year) was observed in summer. For other seasons, AOD was decreasing by -0.022, -0.028 and -0.023 per year (for DJF, MAM, SON, respectively), which is ca. -6% of the AOD per year for each season.
- 10 Over all of China, the AOD decrease was also more pronounced in summer (-0.021, or -7% of AOD, per year). For other seasons, AOD was decreasing by -0.012, -0.014 and -0.011 per year (for DJF, MAM, SON, respectively), which is -4%-of AOD per year in winter and spring and -5% of AOD per year in autumn.

Thus, the AOD changes in P2 in summer contribute most to the AOD annual year-to-year variability. AOD seasonal year-to-year changes are more pronounced over eastern China, which is explained by the uneven regional economic development in China.

- 15 As discussed above, the linear fitting of the whole period WP (1995-2017) does not give significant results, when applied to the annual AOD time series, since annual time series show positive AOD tendency in P1 and negative AOD tendency in P2 in almost all regions (except region 8, as discussed above). For seasonal aggregates, where dAOD has the same sign in P1 and P2 (e.g. spring time series in regions 6 and 7), linear fitting can be applied for the whole period. However, this behavior, where AOD tendency has similar sign in both P1 and P2, is an exception over China and thus, as for yearly aggregates, linear fitting results for the whole period for seasonal aggregates are shown in Table A2 but not discussed in detail. 20

6. Summary and conclusions

With the rapid economic development and further urbanization, the concentration of anthropogenic and natural aerosols accumulate have been increasing in China. However, the emission reduction policies in China have been changing during the last two decades have successfully reduced the concentration of the atmospheric trace gases (van der A et al., 2017), considerably strengthening the

25 emission control after 2011 (Jin et al., 2016). Here we investigate whether the tendencies in the AOD in-during the last two decades are related to emission changes as well as to follow up the pollution control policies in China. The limited lifetime of satellites makes it impossible to follow the AOD changes for-during several decades using time series from only one-with the single instrument. In this paper, we introduced a method to construct a combined-multi-decadal AOD time series by combining data from three different sensors: AATSR-2, AOD time series using the ATSR and MODIS/Terra-sensors, which

30 together cover the period of from 1995 to -2017. The method is based on the ADV and MODIS comparison discussed in Part IPart 1. In brief, (1) ADV and MODIS show similar AOD annual and seasonal spatial and temporal patterns and (2) using AERONET AOD as independent reference data set, ADV is negatively biased, while MODIS is positively biased by about the same amount. against AERONET AOD; in absolute values, the biases are similar to each other. The method was applied pixel-wise to <u>L3</u> annual and seasonal AOD aggregates from ADV and MODIS to construct the combined AOD <u>data settime series</u>.

<u>This combined data set was used to produce The</u>-long-term (1995-2017) annual and seasonal combined-AOD time series-are presented for <u>all</u> China, southeaster (SE)SE China and 10 selected regions. Linear regression <u>has beenwas</u> applied to individual L3 (1°x1°) pixels of the annual and seasonal combined AOD time series to estimate the <u>changes in AOD tendencies</u> over China for three two periods: 1995-2006 (P1) and 2011-2017 (P2), as regarding the changes in the emission reduction policies, and the whole period 1995 2017(WP), when the combined AOD time series is available. Years 2006 and 2011 were identified as pivot points

using a statistical analysis. These pivot points coincide which implementation of the Chinese Five-Years Plans. The length of the periods is too short to estimate AOD trends. Therefore, we discuss AOD tendencies.

- - In P1, associated with the increase of emissions induced by rapid economic development (Jin et al., 2016), AOD increased strongly over the wide industrial areas. For the annual AOD, Pthe positive tendencyies of annual AOD of (0.006 (, or 2% of the AOD averaged over the corresponding period), per year) prevailed across all of mainland China before 2006was observed for all of China. due to emission increases induced by rapid economic development. In P1, AOD was s increasing strongly over the wide industrial areas. Thus, i. In SE China, the annual AOD positive tendency in 1995-2006 was 0.014,
 - or 3%-of AOD, per year-in SE China, reaching maxima (0.020, or 4%-of AOD, per year) in Shanghai and the Pearl River Delta regioains.
 - Negative AOD tendencies (-0.015, or -6% of AOD, per year) were identified across most of China after 2011 in conjunction with effective emission reduction in anthropogenic primary aerosols, SO₂ and NOx (Jin et al., 20172016;, van der A et al., 2017). The air quality mostly improved at the developed regions. Overall, AOD decrease in P2 was 2-3 times stronger than AOD increase in P1 over most of the SE China.- The strongest AOD decrease in P2 is observed in the Chengdu (-0.045, or -8% of AOD, per year) and Zhengzhou (-0.046, or -9% of AOD, per year) areas, while over the North China plane and coastal areas the AOD decrease was <-0.03, or ca -6% per year. In the less populated areas, the AOD decrease was small.</p>
- 25 The AOD tendencies for the whole period 1995-2017 were much less pronounced. The reason for this is that positive AOD tendency has been observed at the beginning of WP (in P1) and negative AOD tendency has been observed at the end of WP (in P2), which partly cancel each other during 1995-2017.
 - Seasonal patterns in the AOD regional long-term tendencies are evident. The contribution of seasonal AOD tendencies to annual tendencies was not equal along the year. While the annual AOD tendency was positive in P1, the AOD tendencies in winter and spring were slightly negative (ca. -0.002, or -1% of AOD, per year) over the most of China during that period. AOD tendencies were positive in summer (0.008, or 2% of AOD, per year) and autumn (0.006, or 6% of AOD, per year) over all mainland China and SE China (0.020, or 4% of AOD, per year and 0.016, or 4% of AOD, per year in summer and autumn, respectively). As in P1, the AOD negative tendencies in P2 were higher compared to other seasons- in summer

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over China (ca. -0.021, or -7% of AOD, per year) and over SE China (ca. -0.048, or -9% of AOD, per year). -In the east, seasonal variations in AOD tendencies were less pronounced.

Thus, in the current study the effect of the changes in the emission regulations policy in China is evident in <u>the</u>AOD decrease after 2011. The effect is more visible in the highly populated and industrialized regions in SE China.

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Data availability

The ATSR data used in this manuscript are publicly available (after registration a password will be issued) at: http://www.icare.univlille1.fr/. MODIS data are publicly available at: https://ladsweb.modaps.eosdis.nasa.gov/. AERONET data are available at AERONET: https://aeronet.gsfc.nasa.gov/

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- 20 AERONET sites used in this study.

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Appendix

Table A1. Comparison of the seasonal and annual ADV, MODIS and Combined AOD with AERONET AOD for different periods: T2 (2000-2011, ADV and MODIS overlapping period), T3 (2011-2017, MODIS), ADV period (1998-2012), MODIS/Terra whole period (2000-2017); number of points (N), correlation coefficient (R), standard deviation (σ) and root meat square error (rms).

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<u>season</u>	<u>collection</u>	<u>N</u>	<u>R</u>	<u>slope</u>	<u>bias</u>	<u>o</u>	<u>rms</u>
<u>T2 (2000-</u>	2011)	_	_	_	_	_	_
_	ADV	<u>71</u>	<u>0,32</u>	<u>0,12</u>	<u>0,2</u>	<u>0,023</u>	<u>0,27</u>
<u>DJF</u>	MODIS	<u>82</u>	<u>0,53</u>	<u>0,17</u>	<u>0,58</u>	<u>0,022</u>	<u>0,2</u>
_	Combined	<u>82</u>	<u>0,54</u>	<u>0,14</u>	<u>0,39</u>	<u>0,019</u>	<u>0,19</u>
_	ADV	<u>81</u>	<u>0,41</u>	<u>0,2</u>	<u>0,26</u>	<u>0,03</u>	<u>0,33</u>
MAM	MODIS	<u>78</u>	<u>0,25</u>	<u>0,42</u>	<u>0,21</u>	<u>0,035</u>	<u>0,31</u>
_	Combined	<u>82</u>	<u>0,4</u>	<u>0,27</u>	<u>0,29</u>	<u>0,031</u>	<u>0,3</u>
_	ADV	<u>76</u>	<u>0,85</u>	<u>-0,03</u>	<u>0,91</u>	<u>0,027</u>	<u>0,24</u>
JJA	MODIS	<u>75</u>	<u>0,84</u>	<u>0,05</u>	<u>0,95</u>	<u>0,029</u>	<u>0,25</u>
_	Combined	<u>76</u>	<u>0,86</u>	<u>0</u>	<u>0,93</u>	<u>0,026</u>	<u>0,23</u>
_	ADV	<u>81</u>	<u>0,57</u>	<u>0,09</u>	<u>0,4</u>	<u>0,023</u>	<u>0,25</u>
<u>SON</u>	MODIS	<u>82</u>	<u>0,67</u>	<u>0,1</u>	<u>0,74</u>	<u>0,023</u>	<u>0,21</u>
_	Combined	<u>82</u>	0,66	<u>0,1</u>	<u>0,57</u>	<u>0,021</u>	<u>0,21</u>
_	ADV	<u>105</u>	0,64	<u>0,09</u>	<u>0,52</u>	<u>0,02</u>	<u>0,25</u>
Year	MODIS	<u>105</u>	<u>0,64</u>	<u>0,14</u>	<u>0,7</u>	<u>0,023</u>	<u>0,23</u>
_	Combined	<u>105</u>	0,65	<u>0,11</u>	<u>0,61</u>	<u>0,021</u>	0,22
<u>T3 (2011-</u>	<u>2017)</u>	_	_	_	_	_	_
DJF	MODIS	<u>52</u>	<u>0,59</u>	<u>0,17</u>	<u>0,8</u>	<u>0,028</u>	0,23
_	Combined	<u>52</u>	<u>0,59</u>	<u>0,15</u>	<u>0,51</u>	<u>0,021</u>	<u>0,15</u>
MAM	MODIS	<u>53</u>	<u>0,49</u>	<u>0,29</u>	<u>0,47</u>	<u>0,032</u>	<u>0,23</u>
_	<u>Combined</u>	<u>58</u>	<u>0,61</u>	<u>0,17</u>	<u>0,5</u>	<u>0,027</u>	<u>0,21</u>
JJA	MODIS	<u>49</u>	<u>0,93</u>	<u>0,03</u>	<u>1,09</u>	<u>0,021</u>	<u>0,16</u>
_	Combined	<u>50</u>	<u>0,94</u>	<u>-0,01</u>	<u>1,04</u>	<u>0,018</u>	<u>0,13</u>
<u>SON</u>	MODIS	<u>44</u>	<u>0,65</u>	<u>0,14</u>	<u>0,77</u>	<u>0,032</u>	<u>0,22</u>
_	Combined	44	<u>0,64</u>	<u>0,12</u>	<u>0,59</u>	<u>0,028</u>	<u>0,18</u>
Year	MODIS	<u>62</u>	<u>0,71</u>	<u>0,1</u>	<u>0,87</u>	<u>0,026</u>	<u>0,21</u>
_	<u>Combined</u>	<u>62</u>	<u>0,71</u>	<u>0,08</u>	<u>0,76</u>	<u>0,023</u>	<u>0,18</u>
ADV perio	<u>od (1998-2012)</u>	_	_	_	_	_	_
<u>DJF</u>	ADV	<u>72</u>	<u>0,33</u>	<u>0,12</u>	<u>0,2</u>	<u>0,023</u>	<u>0,27</u>
	Combined	<u>84</u>	<u>0,54</u>	<u>0,14</u>	<u>0,4</u>	<u>0,018</u>	<u>0,19</u>
MAM	ADV	<u>88</u>	<u>0,39</u>	<u>0,2</u>	<u>0,25</u>	<u>0,029</u>	<u>0,33</u>
_	<u>Combined</u>	<u>89</u>	<u>0,38</u>	<u>0,28</u>	<u>0,28</u>	<u>0,029</u>	<u>0,3</u>
JJA	ADV	<u>81</u>	<u>0,85</u>	-0,04	<u>0,92</u>	<u>0,025</u>	<u>0,24</u>
_	Combined	<u>81</u>	<u>0,86</u>	<u>0</u>	<u>0,94</u>	<u>0,025</u>	0,22

<u>SON</u>	ADV	<u>85</u>	<u>0,57</u>	<u>0,09</u>	<u>0,41</u>	<u>0,022</u>	<u>0,25</u>
-	<u>Combined</u>	<u>86</u>	<u>0,66</u>	<u>0,09</u>	<u>0,58</u>	<u>0,02</u>	<u>0,2</u>
Year	ADV	<u>112</u>	<u>0,62</u>	<u>0,09</u>	<u>0,5</u>	<u>0,02</u>	<u>0,26</u>
-	<u>Combined</u>	<u>112</u>	<u>0,63</u>	<u>0,11</u>	<u>0,59</u>	<u>0,02</u>	<u>0,23</u>
MODIS pe	<u>riod (2000-2017)</u>	_	_	_	_	_	_
DJF	MODIS	<u>124</u>	<u>0,53</u>	<u>0,18</u>	<u>0,61</u>	<u>0,019</u>	<u>0,21</u>
-	<u>Combined</u>	<u>124</u>	<u>0,53</u>	<u>0,15</u>	<u>0,4</u>	<u>0,015</u>	<u>0,18</u>
MAM	MODIS	<u>125</u>	<u>0,31</u>	<u>0,38</u>	<u>0,27</u>	<u>0,026</u>	<u>0,29</u>
-	Combined	<u>133</u>	<u>0,46</u>	<u>0,24</u>	<u>0,34</u>	<u>0,022</u>	<u>0,27</u>
JJA	MODIS	<u>117</u>	<u>0,87</u>	<u>0,05</u>	<u>0,96</u>	<u>0,02</u>	<u>0,22</u>
-	<u>Combined</u>	<u>118</u>	<u>0,88</u>	<u>0,01</u>	<u>0,93</u>	<u>0,018</u>	<u>0,2</u>
<u>SON</u>	MODIS	<u>117</u>	<u>0,65</u>	<u>0,12</u>	<u>0,73</u>	<u>0,02</u>	<u>0,22</u>
-	<u>Combined</u>	<u>117</u>	<u>0,65</u>	<u>0,1</u>	<u>0,57</u>	<u>0,018</u>	<u>0,2</u>
<u>Year</u>	MODIS	<u>158</u>	<u>0,65</u>	<u>0,14</u>	<u>0,7</u>	<u>0,018</u>	<u>0,22</u>
_	Combined	<u>158</u>	0,66	0,11	0,61	0,016	0,21

Table <u>A1A2</u>. AOD tendency (dAOD) per year and statistics for the annual combined AOD time series linear fitting (p-value, bias, slope, and uncertainty and absolute-relative error (rae, %)) for twohree periods: P1 (1995-2006), and P2 (2011-2017) and WP (whole period, 1995-2017) for different regions (r, where region 11 is the whole all mainland China and region 12 is mainland SE China).

-	-	-	P1	-	-	-	=	P2	-	-	-	=	WP	-	=
f	dAOD	-	fit	-	-	dAOD	-	fit	-	-	dAOD	-	fit	-	-
-		p-value	bias	slope	ae,%		p-value	bias	slope	ae,%		p-value	bias	slope	ae,%
4	0,018	0,002	-35,39	0,018	6,82	-0,038	0,003	77,62	-0,038	5,65	0,008	0,004	-15,58	0,008	6,95
2	0,020	0,001	-38,63	0,020	6,69	-0,033	0,008	67,28	-0,033	7,23	0,005	0,049	-10,02	0,005	7,79
3	0,010	0,015	-20,13	0,010	7,45	-0,029	0,010	57,76	-0,029	9,15	0,000	0,833	-0,39	0,000	7,79
4	0,018	0,000	-35,49	0,018	6,41	-0,045	0,001	91,59	-0,045	6,72	0,003	0,277	-5,46	0,003	8,88
5	0,012	0,013	-24,46	0,012	10,33	-0,036	0,000	73,50	-0,036	3,80	0,002	0,291	-4,15	0,002	9,64
6	0,012	0,043	-23,72	0,012	8,63	-0,046	0,000	93,19	-0,046	4 ,64	-0,004	0,192	7,94	-0,004	8,84
7	0,020	0,001	-39,27	0,020	7,13	-0,032	0,006	64,22	-0,032	6,96	0,004	0,159	-6,62	0,004	8,02
8	-0,001	0,427	1,49	-0,001	7,41	-0,002	0,216	4,87	-0,002	9,07	-0,001	0,086	1,14	-0,001	5,02
9	0,010	0,072	-19,68	0,010	17,79	-0,014	0,055	29,37	-0,014	11,31	0,002	0,184	-4,49	0,002	11,20
10	0,007	0,172	-14,49	0,007	17,21	-0,004	0,476	8,87	-0,004	10,02	0,002	0,153	-4,36	0,002	9,91
11	0,006	0,006	-11,61	0,006	6,00	-0,015	0,001	30,20	-0,015	4, 39	0,001	0,397	-1,32	0,001	6,12
12	0,014	0,000	-27,28	0,014	4,81	-0,033	0,000	66,84	-0,033	3,60	0,002	0,259	-4,05	0,002	7,51

_	1	_	<u>P1</u>	_	_	<u>P2</u>							
r	dAOD,	<u>unc</u>	p	<u>re, %</u>	<u>dAOD,</u>	dAOD,	<u>unc</u>	p	<u>re, %</u>	<u>dAOD,</u>			
_	period	-	_	_	year	period	-	_	_	year			
<u>1</u>	<u>0,198</u>	<u>0,051</u>	<u>0,002</u>	<u>6,8</u>	<u>0,018</u>	<u>-0,229</u>	<u>0,037</u>	<u>0,003</u>	<u>5,6</u>	<u>-0,038</u>			
<u>2</u>	<u>0,215</u>	<u>0,046</u>	<u>0,001</u>	<u>6,7</u>	<u>0,020</u>	<u>-0,199</u>	<u>0,041</u>	<u>0,008</u>	<u>7,2</u>	<u>-0,033</u>			
<u>3</u>	<u>0,113</u>	<u>0,042</u>	<u>0,015</u>	<u>7,5</u>	<u>0,010</u>	<u>-0,171</u>	<u>0,038</u>	<u>0,010</u>	<u>9,1</u>	<u>-0,029</u>			

<u>4</u>	<u>0,198</u>	0,042	0,000	<u>6,4</u>	<u>0,018</u>	-0,271	0,034	<u>0,001</u>	<u>6,7</u>	-0,045
<u>5</u>	<u>0,137</u>	<u>0,049</u>	<u>0,013</u>	<u>10,3</u>	<u>0,012</u>	<u>-0,218</u>	<u>0,014</u>	<u>0,000</u>	<u>3,8</u>	<u>-0,036</u>
<u>6</u>	<u>0,134</u>	0,063	<u>0,043</u>	<u>8,6</u>	<u>0,012</u>	-0,276	<u>0,021</u>	<u>0,000</u>	<u>4,6</u>	-0,046
<u>7</u>	<u>0,219</u>	<u>0,047</u>	<u>0,001</u>	<u>7,1</u>	<u>0,020</u>	<u>-0,190</u>	<u>0,036</u>	<u>0,006</u>	<u>7,0</u>	<u>-0,032</u>
<u>8</u>	<u>-0,008</u>	<u>0,010</u>	<u>0,427</u>	<u>7,4</u>	<u>-0,001</u>	<u>-0,014</u>	<u>0,009</u>	<u>0,216</u>	<u>9,1</u>	<u>-0,002</u>
<u>9</u>	<u>0,110</u>	<u>0,059</u>	<u>0,072</u>	<u>17,8</u>	<u>0,010</u>	<u>-0,087</u>	<u>0,031</u>	<u>0,055</u>	<u>11,3</u>	<u>-0,014</u>
<u>10</u>	<u>0,081</u>	<u>0,060</u>	<u>0,172</u>	<u>17,2</u>	<u>0,007</u>	<u>-0,026</u>	<u>0,029</u>	<u>0,476</u>	<u>10,0</u>	-0,004
<u>11</u>	<u>0,065</u>	<u>0,020</u>	<u>0,006</u>	<u>6,0</u>	<u>0,006</u>	<u>-0,089</u>	<u>0,011</u>	<u>0,001</u>	<u>4,4</u>	<u>-0,015</u>
<u>12</u>	<u>0,153</u>	<u>0,027</u>	<u>0,000</u>	<u>4,8</u>	<u>0,014</u>	<u>-0,198</u>	<u>0,016</u>	<u>0,000</u>	<u>3,6</u>	-0,033

10 Table A2A3. AOD tendency (dAOD) per year and statistics for linear fitting (p_-value, bias, slope, absolute-uncertainty (unc) and relative error (rae, %)) for the seasonally combined AOD time series for twohree periods: P1 (1995-2006) and, P2 (2011-2017) and WP (whole period, 1995-2017) for different regions (r, where region 11 is the wholeall mainland China and region 12 is the mainland SE China) and different seasons (s, 1 – winter, 2 – spring, 3 – summer, 4 - autumn).

-	-	-	-	P1	-	-	-	-	<u>P2</u>	-	-	-	-	₩₽	-	-
Ŧ	s	dAOD	-	fit	-	-	dAOD	-	fit	-	-	dAOD	-	fit	-	-
-	-		p-value	bias	slope	ae,%		p-value	bias	slope	ae,%		p-value	bias	slope	ae,%
4	4	0,012	0,150	-24,1	0,012	47,5	-0,024	0,213	4 8,8	-0,024	16,9	0,011	0,001	-21,9	0,011	10,8
4	2	-0,002	0,791	5,1	-0,002	9,7	-0,018	0,261	36,3	-0,018	12,4	-0,003	0,218	7,4	-0,003	6,8
4	3	0,024	0,013	-47,0	0,024	9,4	-0,082	0,017	166,0	-0,082	14,0	0,011	0,045	-20,8	0,011	10,5
4	4	0,015	0,049	-28,5	0,015	12,9	-0,014	0,166	28,0	-0,014	8,6	0,006	0,015	-11,6	0,006	8,1
2	4	0,001	0,819	-1,4	0,001	7,7	-0,027	0,085	54,1	-0,027	13,6	0,004	0,086	-6,8	0,004	7,4
2	2	0,005	0,493	-9,5	0,005	7,9	-0,025	0,034	50,9	-0,025	7,2	-0,003	0,272	6,5	-0,003	6,1
2	3	0,026	0,007	-51,7	0,026	12,1	-0,043	0,190	86,8	-0,043	23,3	0,008	0,086	-15,7	0,008	12,3
2	4	0,019	0,010	-36,6	0,019	11,3	-0,027	0,003	54,5	-0,027	5,8	0,003	0,339	-4,8	0,003	9,6
3	1	0,001	0,779	-1,6	0,001	7,5	-0,023	0,049	4 7,0	-0,023	13,0	-0,001	0,694	1,7	-0,001	7,3
3	2	-0,010	0,153	19,9	-0,010	8,4	-0,032	0,028	64,0	-0,032	10,1	-0,003	0,240	6,3	-0,003	6,8
3	3	0,012	0,039	-22,7	0,012	10,5	-0,029	0,157	59,6	-0,029	23,9	0,000	0,870	1,3	0,000	10,9
3	4	0,017	0,016	-33,8	0,017	13,8	-0,028	0,005	56,6	-0,028	9,1	-0,001	0,861	1,3	-0,001	11,6
4	4	- 0,008	0,245	15,4	-0,008	11,8	-0,019	0,093	38,3	-0,019	11,8	-0,002	0,415	4 ,3	-0,002	8,8
4	2	0,009	0,337	-16,6	0,009	10,2	-0,045	0,005	90,5	-0,045	8,8	-0,004	0,245	7,9	-0,004	7,9
4	3	0,017	0,012	-32,4	0,017	9,9	-0,057	0,076	415,5	-0,057	27,0	0,004	0,357	-6,9	0,004	12,8
4	4	0,026	0,000	-51,7	0,026	9,8	-0,040	0,000	80,8	-0,040	5,4	0,004	0,279	-6,5	0,004	11,0
5	4	0,000	0,947	-0,5	0,000	18,6	-0,029	0,004	58,3	-0,029	12,9	0,001	0,775	-0,9	0,001	13,0
5	2	-0,003	0,530	7,1	-0,003	9,6	-0,025	0,017	51,0	-0,025	11,2	-0,005	0,007	10,5	-0,005	7,0
5	3	0,020	0,040	-38,7	0,020	15,3	-0,057	0,009	115,0	-0,057	13,6	0,005	0,215	-9,5	0,005	12,7
5	4	0,010	0,010	-19,9	0,010	10,5	-0,017	0,013	34,4	-0,017	7,6	0,003	0,041	- 5,5	0,003	7,9
6	1	-0,017	0,161	33,8	-0,017	18,8	-0,026	0,298	52,7	-0,026	27,3	-0,005	0,251	10,5	-0,005	14,4

6	2	-0,019	0,182	39,4	-0,019	14,6	-0,048	0,003	97,2	-0,048	9,0	-0,014	0,001	29,1	-0,014	9,3
6	3	0,019	0,039	-38,2	0,019	14,2	-0,060	0,000	120,5	-0,060	7,2	-0,002	0,641	4 ,0	-0,002	12,2
6	4	0,012	0,047	-22,9	0,012	9,6	-0,032	0,024	65,5	-0,032	13,5	-0,004	0,146	8,4	-0,004	9,7
7	4	-0,005	0,471	11,1	-0,005	12,4	-0,019	0,232	37,7	-0,019	16,8	-0,003	0,219	7,0	-0,003	9,1
7	2	-0,006	0,395	11,9	-0,006	7,5	-0,025	0,231	51,4	-0,025	13,1	0,007	0,040	-13,7	0,007	7,5
7	3	0,026	0,012	-51,6	0,026	16,0	-0,033	0,103	66,7	-0,033	18,6	0,001	0,703	-2,3	0,001	12,7
7	4	0,032	0,001	-63,6	0,032	12,8	-0,035	0,008	70,6	-0,035	9,7	0,002	0,690	-2,5	0,002	13,1
8	4	0,002	0,123	-4,0	0,002	14,0	0,001	0,657	-1,3	0,001	12,7	0,000	0,942	0,1	0,000	9,3
8	2	-0,001	0,551	1,8	-0,001	6,7	-0,001	0,505	1,8	-0,001	4,1	0,000	0,918	0,1	0,000	5,1
8	3	-0,001	0,377	1,5	-0,001	6,3	0,001	0,888	-0,9	0,001	15,5	0,000	0,488	-0,4	0,000	5,6
8	4	-0,003	0,016	5,4	-0,003	10,0	-0,003	0,309	5,2	-0,003	16,4	-0,001	0,002	2,6	-0,001	7,8
9	4	-0,039	0,251	69,5	-0,035	115,3	0,001	0,917	-1,6	0,001	30,1	-0,011	0,101	22,0	-0,011	63,9
9	2	-0,047	0,132	94,0	-0,047	39,4	-0,029	0,065	58,3	-0,029	14,2	-0,014	0,058	27,9	-0,014	20,9
9	3	0,007	0,432	-14,5	0,007	28,6	-0,009	0,505	18,9	-0,009	25,6	0,000	0,954	0,6	0,000	16,6
9	4	0,002	0,403	-4,5	0,002	18,7	-0,013	0,127	26,4	-0,013	23,8	0,002	0,060	-4,3	0,002	13,7
10	4	-0,028	0,200	4 9,8	-0,025	68,3	-0,013	0,337	27,0	-0,013	33,6	-0,006	0,236	10,8	-0,005	33,5
10	2	0,004	0,793	-6,7	0,004	28,8	0,001	0,852	-2,2	0,001	11,3	-0,003	0,405	5,8	-0,003	15,7
10	3	0,003	0,544	-6,2	0,003	15,0	-0,016	0,232	33,3	-0,016	18,2	0,002	0,272	-3,9	0,002	40,1
10	4	0,005	0,264	-8,9	0,005	21,4	-0,003	0,349	5,8	-0,003	6,6	0,004	0,003	-7,1	0,004	10,6
-11	4	-0,001	0,556	2,9	-0,001	7,4	-0,012	0,041	23,9	-0,012	8,9	0,001	0,606	-0,7	0,001	6,0
-11	2	-0,003	0,555	6,5	-0,003	10,5	-0,014	0,030	28,7	-0,014	7,8	-0,003	0,020	7,2	-0,003	6,2
-11	3	0,008	0,006	-14,7	0,008	6,6	-0,021	0,036	42,2	-0,021	12,9	0,001	0,477	-1,7	0,001	7,9
-11	4	0,006	0,001	-11,5	0,006	6,1	-0,011	0,007	21,8	-0,011	6,1	0,001	0,130	-2,1	0,001	6,2
12	4	0,001	0,642	-1,9	0,001	5,6	-0,022	0,024	44,1	-0,022	9,7	0,002	0,232	-3,2	0,002	6,6
12	2	-0,003	0,476	7,0	-0,003	6,2	-0,028	0,012	55,9	-0,028	7,9	-0,004	0,031	8,7	-0,004	5,4
12	3	0,020	0,001	-39,0	0,020	7,5	-0,048	0,012	97,1	-0,048	13,2	0,004	0,232	-7,3	0,004	10,6
12	4	0,016	0,000	-31,5	0,016	7,4	-0,023	0,000	4 7,2	-0,023	4,0	0,002	0,346	-3,2	0,002	8, 4

_	_	_	_	<u>P1</u>	_	_	-	_	<u>P2</u>	_	_
<u>r</u>	<u>s</u>	dAOD,	<u>unc</u>	<u>p</u>	<u>re, %</u>	dAOD,	dAOD,	unc	<u>p</u>	<u>re, %</u>	dAOD,
_	_	period	_	_	_	year	period	_	_	_	year
<u>1</u>	<u>SON</u>	<u>0,159</u>	<u>0,077</u>	<u>0,049</u>	<u>12,9</u>	<u>0,015</u>	<u>-0,082</u>	<u>0,045</u>	<u>0,166</u>	<u>8,6</u>	<u>-0,014</u>
<u>2</u>	DJF	<u>0,009</u>	<u>0,041</u>	<u>0,819</u>	<u>7,7</u>	<u>0,001</u>	<u>-0,160</u>	<u>0,066</u>	<u>0,085</u>	<u>13,6</u>	<u>-0,027</u>
<u>2</u>	MAM	<u>0,046</u>	<u>0,064</u>	<u>0,493</u>	<u>7,9</u>	<u>0,005</u>	<u>-0,150</u>	<u>0,045</u>	<u>0,034</u>	<u>7,2</u>	<u>-0,025</u>
<u>2</u>	JJA	<u>0,288</u>	<u>0,092</u>	<u>0,007</u>	<u>12,1</u>	<u>0,026</u>	<u>-0,257</u>	<u>0,149</u>	<u>0,190</u>	<u>23,3</u>	<u>-0,043</u>
<u>2</u>	<u>SON</u>	0,204	<u>0,070</u>	<u>0,010</u>	<u>11,3</u>	<u>0,019</u>	<u>-0,161</u>	<u>0,027</u>	<u>0,003</u>	<u>5,8</u>	<u>-0,027</u>
<u>3</u>	DJF	0,010	<u>0,036</u>	<u>0,779</u>	<u>7,5</u>	<u>0,001</u>	<u>-0,139</u>	<u>0,047</u>	<u>0,049</u>	<u>13,0</u>	-0,023
<u>3</u>	MAM	<u>-0,087</u>	<u>0,056</u>	<u>0,153</u>	<u>8,4</u>	<u>-0,010</u>	<u>-0,189</u>	<u>0,054</u>	<u>0,028</u>	<u>10,1</u>	<u>-0,032</u>
<u>3</u>	<u>JJA</u>	<u>0,127</u>	<u>0,058</u>	<u>0,039</u>	<u>10,5</u>	<u>0,012</u>	<u>-0,176</u>	<u>0,093</u>	<u>0,157</u>	<u>23,9</u>	<u>-0,029</u>
<u>3</u>	<u>SON</u>	<u>0,188</u>	<u>0,070</u>	<u>0,016</u>	<u>13,8</u>	<u>0,017</u>	<u>-0,168</u>	<u>0,032</u>	<u>0,005</u>	<u>9,1</u>	<u>-0,028</u>
<u>4</u>	<u>DJF</u>	<u>-0,075</u>	<u>0,063</u>	<u>0,245</u>	<u>11,8</u>	<u>-0,008</u>	<u>-0,113</u>	<u>0,048</u>	<u>0,093</u>	<u>11,8</u>	<u>-0,019</u>
<u>4</u>	MAM	<u>0,078</u>	<u>0,077</u>	<u>0,337</u>	<u>10,2</u>	0,009	-0,268	<u>0,050</u>	<u>0,005</u>	<u>8,8</u>	-0,045
<u>4</u>	JJA	<u>0,181</u>	<u>0,064</u>	<u>0,012</u>	<u>9,9</u>	<u>0,017</u>	<u>-0,343</u>	<u>0,135</u>	<u>0,076</u>	<u>27,0</u>	<u>-0,057</u>
<u>4</u>	<u>SON</u>	<u>0,287</u>	<u>0,061</u>	<u>0,000</u>	<u>9,8</u>	<u>0,026</u>	<u>-0,239</u>	<u>0,026</u>	<u>0,000</u>	<u>5,4</u>	<u>-0,040</u>
<u>5</u>	<u>DJF</u>	<u>0,004</u>	<u>0,056</u>	<u>0,947</u>	<u>18,6</u>	<u>0,000</u>	<u>-0,173</u>	<u>0,031</u>	<u>0,004</u>	<u>12,9</u>	<u>-0,029</u>
<u>5</u>	MAM	<u>-0,030</u>	<u>0,046</u>	<u>0,530</u>	<u>9,6</u>	<u>-0,003</u>	<u>-0,151</u>	<u>0,038</u>	<u>0,017</u>	<u>11,2</u>	<u>-0,025</u>
<u>5</u>	<u>JJA</u>	<u>0,216</u>	<u>0,099</u>	<u>0,040</u>	<u>15,3</u>	<u>0,020</u>	<u>-0,341</u>	<u>0,072</u>	<u>0,009</u>	<u>13,6</u>	<u>-0,057</u>
<u>5</u>	<u>SON</u>	<u>0,111</u>	<u>0,038</u>	<u>0,010</u>	<u>10,5</u>	0,010	-0,102	<u>0,024</u>	<u>0,013</u>	7,6	-0,017
<u>6</u>	DJF	<u>-0,167</u>	<u>0,114</u>	<u>0,161</u>	<u>18,8</u>	<u>-0,017</u>	<u>-0,156</u>	<u>0,118</u>	<u>0,298</u>	<u>27,3</u>	<u>-0,026</u>
<u>6</u>	MAM	<u>-0,174</u>	<u>0,120</u>	<u>0,182</u>	<u>14,6</u>	<u>-0,019</u>	<u>-0,288</u>	<u>0,047</u>	0,003	<u>9,0</u>	<u>-0,048</u>
<u>6</u>	JJA	0,213	0,098	0,039	<u>14,2</u>	0,019	-0,358	0,032	0,000	7,2	-0,060
<u>6</u>	<u>SON</u>	<u>0,129</u>	<u>0,062</u>	<u>0,047</u>	<u>9,6</u>	<u>0,012</u>	-0,194	<u>0,054</u>	<u>0,024</u>	<u>13,5</u>	-0,032

<u>7</u>	DJF	<u>-0,053</u>	<u>0,074</u>	<u>0,471</u>	<u>12,4</u>	<u>-0,005</u>	<u>-0,111</u>	<u>0,072</u>	<u>0,232</u>	<u>16,8</u>	<u>-0,019</u>
<u>7</u>	MAM	<u>-0,051</u>	<u>0,057</u>	<u>0,395</u>	<u>7,5</u>	<u>-0,006</u>	<u>-0,151</u>	<u>0,098</u>	<u>0,231</u>	<u>13,1</u>	<u>-0,025</u>
<u>7</u>	JJA	<u>0,287</u>	<u>0,102</u>	0,012	<u>16,0</u>	<u>0,026</u>	-0,197	0,087	0,103	<u>18,6</u>	-0,033
<u>7</u>	<u>SON</u>	<u>0,353</u>	<u>0,079</u>	<u>0,001</u>	<u>12,8</u>	<u>0,032</u>	<u>-0,209</u>	<u>0,043</u>	<u>0,008</u>	<u>9,7</u>	<u>-0,035</u>
<u>8</u>	DJF	<u>0,018</u>	<u>0,011</u>	<u>0,123</u>	<u>14,0</u>	<u>0,002</u>	<u>0,004</u>	<u>0,008</u>	<u>0,657</u>	<u>12,7</u>	<u>0,001</u>
<u>8</u>	MAM	<u>-0,007</u>	<u>0,012</u>	<u>0,551</u>	<u>6,7</u>	<u>-0,001</u>	<u>-0,005</u>	<u>0,006</u>	<u>0,505</u>	<u>4,1</u>	<u>-0,001</u>
<u>8</u>	JJA	-0,008	<u>0,009</u>	<u>0,377</u>	<u>6,3</u>	<u>-0,001</u>	<u>0,003</u>	<u>0,018</u>	<u>0,888</u>	<u>15,5</u>	<u>0,001</u>
<u>8</u>	<u>SON</u>	<u>-0,029</u>	<u>0,011</u>	<u>0,016</u>	<u>10,0</u>	<u>-0,003</u>	<u>-0,015</u>	<u>0,012</u>	<u>0,309</u>	<u>16,4</u>	<u>-0,003</u>
<u>9</u>	<u>DJF</u>	<u>-0,312</u>	<u>0,247</u>	<u>0,251</u>	<u>115,3</u>	<u>-0,039</u>	<u>0,005</u>	<u>0,043</u>	<u>0,917</u>	<u>30,1</u>	<u>0,001</u>
<u>9</u>	MAM	<u>-0,420</u>	<u>0,253</u>	<u>0,132</u>	<u>39,4</u>	<u>-0,047</u>	<u>-0,172</u>	<u>0,065</u>	<u>0,065</u>	<u>14,2</u>	<u>-0,029</u>
<u>9</u>	<u>AII</u>	<u>0,082</u>	<u>0,108</u>	<u>0,432</u>	<u>28,6</u>	<u>0,007</u>	<u>-0,055</u>	<u>0,068</u>	<u>0,505</u>	<u>25,6</u>	<u>-0,009</u>
<u>9</u>	<u>SON</u>	<u>0,025</u>	<u>0,032</u>	<u>0,403</u>	<u>18,7</u>	<u>0,002</u>	<u>-0,078</u>	<u>0,038</u>	<u>0,127</u>	<u>23,8</u>	-0,013
<u>10</u>	DJF	-0,223	<u>0,157</u>	<u>0,200</u>	<u>68,3</u>	-0,028	<u>-0,080</u>	<u>0,066</u>	<u>0,337</u>	<u>33,6</u>	<u>-0,013</u>
<u>10</u>	MAM	<u>0,032</u>	<u>0,118</u>	<u>0,793</u>	<u>28,8</u>	<u>0,004</u>	<u>0,007</u>	<u>0,033</u>	<u>0,852</u>	<u>11,3</u>	<u>0,001</u>
<u>10</u>	JJA	<u>0,036</u>	<u>0,062</u>	<u>0,544</u>	<u>15,0</u>	<u>0,003</u>	<u>-0,098</u>	<u>0,064</u>	<u>0,232</u>	<u>18,2</u>	<u>-0,016</u>
<u>10</u>	<u>SON</u>	<u>0,050</u>	<u>0,046</u>	<u>0,264</u>	<u>21,4</u>	<u>0,005</u>	<u>-0,017</u>	<u>0,014</u>	<u>0,349</u>	<u>6,6</u>	<u>-0,003</u>
<u>11</u>	DJF	<u>-0,013</u>	<u>0,023</u>	<u>0,556</u>	<u>7,4</u>	<u>-0,001</u>	<u>-0,071</u>	<u>0,023</u>	<u>0,041</u>	<u>8,9</u>	<u>-0,012</u>
<u>11</u>	MAM	-0,028	<u>0,045</u>	<u>0,555</u>	<u>10,5</u>	<u>-0,003</u>	<u>-0,084</u>	<u>0,025</u>	<u>0,030</u>	<u>7,8</u>	-0,014
<u>11</u>	<u>JJA</u>	<u>0,083</u>	<u>0,026</u>	<u>0,006</u>	<u>6,6</u>	<u>0,008</u>	<u>-0,125</u>	<u>0,039</u>	<u>0,036</u>	<u>12,9</u>	<u>-0,021</u>
<u>11</u>	<u>SON</u>	<u>0,065</u>	<u>0,016</u>	<u>0,001</u>	<u>6,1</u>	<u>0,006</u>	<u>-0,064</u>	<u>0,013</u>	<u>0,007</u>	<u>6,1</u>	<u>-0,011</u>
<u>12</u>	DJF	<u>0,011</u>	<u>0,024</u>	<u>0,642</u>	<u>5,6</u>	<u>0,001</u>	<u>-0,130</u>	<u>0,036</u>	<u>0,024</u>	<u>9,7</u>	-0,022
<u>12</u>	MAM	-0,029	<u>0,039</u>	<u>0,476</u>	<u>6,2</u>	<u>-0,003</u>	<u>-0,165</u>	<u>0,038</u>	<u>0,012</u>	<u>7,9</u>	<u>-0,028</u>
<u>12</u>	JJA	<u>0,217</u>	<u>0,048</u>	<u>0,001</u>	<u>7,5</u>	<u>0,020</u>	<u>-0,288</u>	<u>0,066</u>	<u>0,012</u>	<u>13,2</u>	<u>-0,048</u>
<u>12</u>	<u>SON</u>	<u>0,175</u>	<u>0,036</u>	<u>0,000</u>	<u>7,4</u>	<u>0,016</u>	-0,140	<u>0,015</u>	<u>0,000</u>	<u>4,0</u>	-0,023