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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temporal and spatial analysis of ozone concentrations in Europe based on time scale decomposition and a multi-clustering approach" *by* Eirini Boleti et al.

Anonymous Referee #1

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Review of Boleti et al. (ACPD, 2019)

Boleti et al. have used a time series decomposition methodology introduced in their previous papers to extract the long-term, seasonal and short-term components of ozone time series at around 300 measurement sites in Europe. Then they have applied a clustering algorithm separately on the long-term and seasonal components to get a two-dimensional classification of sites, according to the site type (proximity to emission sources) and to the regional characteristics (meteorological influence). Through the combination of such techniques they have gone a step forward compared to previous analyses that resulted in regional site (Carro-Calvo et al., 2017) or site type (Lyapina et

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al., 2016) classifications. In addition, grouping the sites according to the two categories as done here simplifies the interpretation of long-term ozone trends.

The manuscript also includes some other powerful analyses, such as the application of a meteorological adjustment technique which has allowed to obtain significantly negative trends of summer peak ozone concentrations at many more sites than in previous trend assessments (e.g. Fleming et al, 2018). Furthermore, through the examination of the seasonal component they document the distinct behavior of some clusters (e.g. maximum ozone occurring earlier in the year over northern Europe than over the Po Valley) as well as a reduction of the amplitude of such cycles and a shift of the day with ozone maximum.

The manuscript represents a substantial contribution to the field and considers related work by including appropriate references. I indeed find it very appropriate for publication in Atmos. Chem. Phys, but at the same time think it should substantially be improved. I have three major concerns. Two of them are related to (i) the choice of daily O3 (instead of MDA8 O3) for the main analyses presented in the manuscript, which has not been justified by the authors, and to (ii) the disconnection between the main text and large parts of the supplementary material (see main comments). My third concern is that the authors should spend time on improving some parts of the manuscript, as seen from the large number of comments included below. I think the manuscript contains a considerable number of inaccuracies, but will fully support the publication once the authors have addressed these comments.

MAIN COMMENTS

(1) The authors address the spatiotemporal variability of daily mean O3 in the main text and leave MDA8 O3 for the supplement. In particular, it is a bit surprising that the daily mean concentrations during the extended summer season are used in section 4.3 (Trends of peak O3 concentrations). Wouldn't it have been more appropriate to use MDA8 O3 at least for that section to focus on the times of the day with the highest

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ozone concentrations?

I am not against this choice, but think that the authors should at least justify it. Are they using daily O3 because that simplifies the comparison of most of their results with those of other studies? If that was the case, I would understand that they have preferred sticking to daily O3 in all sections of the main text, just for consistency. Or is there any other reason?

(2) Overall, the main text and the supplement look like two completely separated pieces of work which are not properly linked. The Supplementary Material includes 5 sections and 16 additional Figures, but most of them are neither explained nor referred to from the main text. This is very unpleasant for the reader, who has to look for the appropriate sections and figures in the supplement. Bottom line: the authors should explicitly mention which section/figure of the supplement they are referring to; at the same time, they should not include analyses in the supplement if they do not refer to them from the main text. Here are just some examples:

* Lines 8-11 of page 6: “In addition, a Silhouette width (Sw) analysis is performed to assess the goodness of the clustering (Rousseeuw, 1987). More details about the number of clusters, the goodness of the clustering and the Sw are provided in the supplementary material”. Need to refer to some specific sections? Maybe S1-S2?

* Lines 5-6 of page 8: “Here, we present the results of the daily mean LT(t)- and S(t)-clustering; results for the W(t)-clustering and the cluster analysis based on the MDA8 are provided in the supplementary material”. Which sections and/or figures of the supplement you are referring to?

* Line 19 of page 9: “The LT(t) signal as derived from the daily mean and MDA8 O3 observations increases” could be changed to The “LT(t) signal as derived from the daily mean (Fig. 3) and MDA8 O3 (Fig. S9) observations increases”.

* The results from Sections S3 and S4 (clusters and trends for MDA8 O3) are not

very useful for the reader because there are hardly any specific comments about them in the main text. For instance, are the trends of daily O3 (main text) and MDA8 O3 (supplement) similar? Are the clusters of their L(t), S(t) and W(t) components overall consistent? The authors have two options: either linking the supplement and the main text much better than done now or removing many things from the supplement (e.g. focus only on daily O3 or on MDA8 O3, see previous main comment). I simply think that so much information without some proper explanations in the main text distracts the reader.

SPECIFIC COMMENTS

(1) There are some parts of Section 3 (Methodology) which need further explanations:

1.1. Additional details on the time scale decomposition should be given. For instance, the text around lines 13-15 of page 5 is not very complete: "By adding together the IMFs with frequencies between around 40 days and 3 years we obtain the seasonal variation of O3 (S(t) = c7 + ... + c10) and by adding the frequencies that are smaller than 40 days the short-term variation is acquired (W (t) = c1 + ... + c6)".

First of all, according to Eq (2), the IMFs (Ci) are time dependent. So I believe it should be "S(t) = c7 (t) + ... + c10 (t)" and "W (t) = c1(t) + ... + c6(t)".

The authors should explain where this decomposition (e.g. c7 to c10) and the corresponding time scales (e.g. 40 days to 3 years) come from. If this comes from the previous analyses by Boleti et al (2018) they should explicitly state that.

1.2. The description of the partitioning around medoids (PAM) clustering algorithm used in this study is hard to understand.

For instance, around line 21 of page 5: "PAM is more robust than k-means, because it minimizes the sum of dissimilarities instead of the sum of squared euclidean distances . . . Initially, k clusters are generated randomly and the empirical means mk of the euclidean distance between their data points are calculated . . .". First the authors say that

PAM does not minimize the sum of squared Euclidean distances but then they mention "euclidean distance" when they refer to mk. I do not get it. By the way, I think it should be "Euclidean" instead of "euclidean".

Around lines 3-4 of page 6: "To identify the optimal number of clusters the k-means algorithm is iteratively executed for a range of k values . . .". Now, you are referring to k-means instead of to PAM. Can you please explain all this better? From the present text it is not easy to understand what is different in k-means and PAM.

1.3. Meteorological adjustment (Section 3.4). The authors use GAMS models to fit ozone on a number of variables (eq. 6). Then they follow Barmpadimos et al (2011) to calculate meteo-adjusted ozone as a function of the temporal trend and the residual from the models (eq. 7). Can you please briefly mention how the variable selection is done? Using step-wise regression like in Barmpadimos' work? And what is the overall performance of the meteo adjustement? Similar to that found by previous papers by the same authors for Swiss sites?

(2) The authors should provide further details about the choice, importance and characteristics of the Po Valley cluster (derived from the seasonal component of daily O3, see e.g. Figure 4). Some questions:

2.1. According to that figure and Table 1, the cluster only includes 4 sites. This is too little compared to the other clusters and therefore needs some justification. Would have this cluster appeared if the authors had kept only k=4 instead of k=5 clusters? Even if that was not the case, I understand that it might be appropriate to retain this cluster if the characteristics of this region are very different from those in the surroundings (e.g. elevated emissions and confinement of pollution within a basin with little ventilation, distinct annual cycle as seen from Figure 5).

2.2. around line 29 of page 9: "The sites in "PoValley" display the most pronounced S(t), mainly due to the Mediterranean weather conditions, e.g. high temperatures. At the same time high NOx and VOC emissions in this region leads to higher O3 concen-

trations". I am not convinced at all with this statement. Note that the amplitude of the S(t) component is remarkably wider both for the Po Valley and the Central North cluster compared to the others (Figure 4). I am surprised at the results for the Central North cluster, where I would expect average ozone concentrations during the warm season (but not in the colder months) to be clearly below those in the Po valley. The authors should explicitly mention this similarity between two apparently very different regions and, if possible, explain why this happens. In other words, are there any reasons why the impact of meteorology and emissions on ozone presents stronger seasonality in these two regions than in others?

In addition, I would remove "Mediterranean weather conditions, e.g. high temperatures", which I find quite vague. I think the expression "Mediterranean weather conditions" is much more appropriate for the coastal sites in Spain, southeastern France and in the proximity of Rome (see Figure 1). I am not sure that "e.g. high temperatures" is appropriate either here because this analysis includes ozone data in all seasons.

(3) In the long paragraph between lines 4-21 of page 10 the authors compare the results to those of previous classifications, namely Carro-Calvo et al. (2017) and Lyapina et al. (2016). See comments:

3.1. The comparison of the results of the S(t) clustering to those by Carro-Calvo is probably too exhaustive. I would simplify it, but this is up to the authors to decide whether they want to do that. Rather than mentioning every single regional difference arising from the comparison of both classifications, I would instead list all the possible reasons why the results of both classifications ae expected to differ. Only some of those reasons are mentioned in the text. Basically, Carro-Calvo used a MDA8 O3 gridded dataset considering only the summer months, while daily O3 at specific sites during the whole year is used here. In addition, Carro-Calvo applied k-means on normalized anomalies while the spatial classification presented here is based on the seasonal component. Finally, the authors are right to indicate that some of the clusters of Carro-Calvo et al. (2017) do not appear here because the former study used gridded data

over locations with few observations, but this explanation is not complete. Note that the final number of clusters will depend on the a priori choices made (e.g. decisions on the number of clusters based on the explained variance achieved, intra-cluster variance or RMSE, minimizing correlations among different clusters, silhouette width, and so on).

3.2. I feel the comparison of the results from the L(t) clustering to those of Lyapina et al. (2016) would benefit from some additional explanations. That work performed two cluster analyses (CA). The first CA used absolute mixing ratio values and resulted in 5 clusters (Table 2 of that paper), while the second CA used normalized mixing rations and yielded 4 clusters (Table 3 of that paper). As it is not straightforward to summarize the description of the clusters in those tables, one could just select one of them (e.g. the first one) and provide some simple explanations. For instance, one could indicate that the results from this study are similar to those of a classification by Lyapina et al. (2016) who found 5 clusters of different type, ranging from urban traffic (equivalent to the "highly polluted" reported here) to rural background.

3.3. It is very good that the authors have acknowledged previous work and compared their results to those studies. Apart from that, either here or somewhere else in the paper, I would emphasize the strength of this work: they authors have clearly gone a step forward compared to those studies because they have provided a two-dimensional classification.

(4) Figure 5 (Annual cycle of daily mean O3 S(t) for the daily mean S(t) clusters) appears on page 12, but I think it is not referenced to from the main text. The figure should be moved to another part of the text (Section 4.4. Ozone seasonal cycles), which would affect the numbering of other figures. Then, in section 4.4, it would be good to mention some of the main features seen from the S(t) component of daily O3 in that figure. For instance, the figure nicely shows that the ozone maxima occurs in summer for the Po Valley cluster and much earlier in the year in the North cluster. This is consistent with previous studies that have reported that both the highest average ozone concentrations and extreme ozone episodes tend to occur over central/southern

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Europe during summer and over northern Europe in spring (see e.g. Figs 1 of both Schnell et al., 2015 and Ordóñez et al., 2017). Finally, I would explicitly mention the days of the ozone maxima in each cluster when commenting the trend of DoMax in Table 2.

(5) Section 4.2 is on trends of daily mean ozone, but Figure 7 at the end of that section shows results of MDA8 LT for Mace Head. Why do you use MDA8 instead of daily O3 for that particular figure? Is it just to compare the results with those of Derwent's papers (see first paragraph of page 14)?

(6) As seen from the first paragraph of section 4.3 (trends of peak O3) the main result from that section is that, unlike previous studies like that of Flemming et al. (2018), the meteorological adjustment results in significantly negative trends at many sites. That is a very nice result, but I am not fully convinced with all the interpretations of the trends in the following paragraph. For instance, around lines 17-20 of page 14: "in the "BAC" cluster (especially the "West" cluster) the decrease of MTDM was not so pronounced, likely due to the increase of hemispheric transport of O3 in Europe (Derwent et al., 2007; Vingarzan, 2004)". However, those papers roughly cover the first halve of the period of analysis, where I agree that might have been the case (see e.g. Figure 7 for a different metric at Mace Head). Moreover, a few lines below (lines 1-3 of page 15) they claim that there might be some connection between the industrialization of Eastern Europe and the trends in some clusters (lines 1-3 of page 15).

I admit that these interpretations are plausible and that the authors have been reasonably careful with their statements, but I would add a short sentence to mention that some more dedicated analyses (e.g. modelling studies) would be needed to investigate the reasons for such trends. I fully understand that such analyses are out of the scope of this paper.

(7) I also like the idea of examining the seasonal cycles of O3 in Section 4.4 and the results presented there are relevant. However, I am not convinced about some of the

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explanations given there as there are some inaccuracies. In addition, I am not happy at all with the writing and believe that this section has been written in a rush. There are so many inaccuracies and corrections to make (some of them included in the technical corrections section) that it very hard to focus on the science. Examples:

* Lines 17-19 of page 16: "The increase in the S min (t) may be partially due to the . . . and probably due to the increased influx of O3 towards north and northwest Europe and more cyclonic activity in the North Atlantic during winter as well (Pausata et al., 2012)". Apart from improving the writing (too many "ands" within the same sentence), I am not convinced at the explanations regarding Pausata's paper. What you do mean by increased influx and cyclonic activity? Are such things really mentioned that way in that paper? If so please explain this better. As far as I remember, that work simply suggested that the increasing baseline ozone in western and northern Europe during the 1990s could be associated with a prevailing NAO+ phase during that period.

* Lines 24-27 of page 16: "The observed shift of the day of seasonal maximum might be linked to the increase of emissions in East Asia that have contributed to increased transport of air pollution to middle-and northern latitudes (Zhang et al., 2016) where the effect on O3 is probably greater due to greater convection, reaction rates and NOx sensitivity [some refs.]. . .". Need to completely rewrite this sentence because it is hard to understand. I assume that the strong convection takes place in East/Southeast Asia instead of at mid/north latitudes as it reads now from this sentence. In addition, the word "greater" is repeated within the same line.

* Line 31 – 33: "The positive phase of the NAO leads to increased O3 concentrations in Europe through higher westerly winds across the North Atlantic, and enhanced transport of air pollutants from North America to Europe (Creilson et al., 2003)." All this looks a bit redundant. Do you simply mean that "The positive phase of the NAO leads to increased O3 concentrations in Europe through enhanced transport of ozone and precursors across the North Atlantic from North America to Europe (Creilson et al., 2003)"?

TECNICAL COMMENTS AND CORRECTIONS

* Line 3 of page 1 (abstract): “the effect of these reductions on ozone is investigated by analyzing surface measurements of ozone”. Change the second “ozone” to “this pollutant” just to avoid redundancies.

* Lines 13-14 of page 1: “The effect of hemispheric transport of ozone can be seen either in regions affected by synoptic patterns in the northern Atlantic or at sites located at remote high altitude locations”. I do not consider this as an appropriate sentence for the abstract. The manuscript includes some references on the impact of long-range transport and changing weather patterns (e.g. impact of the NAO), but it does not provide any supportive evidence of the relevance of such processes.

* Lines 17-18 of page 1: “while seasonal cycle trends and changes in the sensitivity of ozone to temperature are driven by regional climatic conditions”. I would tone down this statement. Honestly, I do not think that the manuscript proves that this impact is larger than that of the varying rates of ozone precursor emission reductions over the different regions.

* Last line of page 2 (Introduction): “For instance, in the U.S., O3 climate penalty – defined as the slope of the O3 versus temperature relationship – dropped from 3.2 ppbv/C before 2002 to 2.2 ppbv/C after 2002 as a result of NOx emission reductions (Bloomer et al., 2009)”. One could add a reference to Colette et al. (2015), who analysed chemistry-transport and climate-chemistry model projections to asses the impact of climate change on this climate penalty over Europe by the turn of the century.

* First line of page 4: “during May and September” –> between May and September

* 5th line of page 4: Did you use surface pressure or sea level pressure (SLP)?

* Line 21 of page 5: Move “e.g.” to the beginning in “(Lyapina et al., 2016, e.g.)”.

* Line 17 of page 6: where yd (t) the de-seasonalized –> where yd (t) is the de-seasonalized

* Line 19 of page 6: “of the” can be removed from “de-seasonalized concentrations of the yd (t)”.

* Line 25 of page 6: “because” is repeated within the same sentence.

* Lines 4-5 of page 7: “For the GAMs, the following meteorological variables were used”. I would remove “meteorological”. Reason: the Julian day, which is not a meteorological variable, is also included in the model.

* Line 6 of page 7: Again “daily mean surface pressure”. Do you mean SLP?

* Line 7 of page 7: No need to spell out CAPE again.

* Line 8 of page 9: “mostly located at higher altitudes”. Higher than what? “Higher” could be changed to something like “relatively high” or “elevated”.

* Lines 9-10 of page 11: “the positive trends can be partly explained by . . . originating from the diesel vehicles”. Change to “the positive trends could partly be explained by . . . originating from the proliferation of diesel vehicles”

* Line 6 of page 12: “rural sites and small and non-significant” –> “rural sites as well as small and non-significant”

* Figure 6: Please indicate what the box-plots indicate (i.e. median, edges of the boxes: 25-27th percentiles, whiskers related to interquartile range or to some specific percentiles, etc.).

* Lines 7-8 of page 13: “Trends were estimated 0.08 ppb/year [0.06,0.1] for the first period and -0.04 ppb/year [-0.09,0.02] for the second period”. Lines 1-2 of page 14: “Derwent et al. (2018) have found an increase of 0.34 $\pm$ 0.07 ppb/year with a deceleration rate after 2007 of -0.0225$\pm$ 0.008 ppb/year”. If possible, indicate if the uncertainty estimates correspond to the 95% confidence intervals or to something else. There are other parts of the text where this is not clear.

* Section 4.2 is on the trends of daily mean ozone, but Figure 7 at the end of that

section shows results of MDA8 LT for Mace Head. The results look convincing, but why do the authors use MDA8 instead of daily O3 for that particular figure? Is this just to compare the results with those of Derwent's papers (see first paragraph of page 14)?

* Lines 6-7 of page 14: The word "increased" can be removed from both lines.

* Lines 1 and 5 of page 16: "central Northeast Germany" and "central Northeast region". Do you mean "central and northeast"?

* Line 28 of page 16: "meteorological factors have affected" –> "meteorological factors may have affected".

* Line 30 of page 16: Add space before while in "data,while".

* Line 34 of page 16: "resulted to"–> "resulted in" (this should be changed somewhere in the supplement too).

* Caption of Figure 9 on page 17: Change "pm" to "+/-" in "average pm the standard deviation".

* Line 2 of page 17: "more increased" –> "increased"

* Lines between pages 17 and 18: The early spring maximum in the "North" sites in April can be explained by higher NOx that is released from PAN and . . .". What do you mean by "higher NOx? Higher than what? Do you mean something like elevated NOx? By the way, why don't you refer to Figure 5 here (see comment above)?

* Table 2 on page 18: According to the methods section, one should write "SDoM". Need to change that in the caption and header of last column.

* Lines 6-9 of page 19: "At PoValley sites the decrease was more pronounced (-0.083 ppb/K/year). At the same time the average correlation between O3 and temperature is the highest compared to the other regions, because of large reductions of precursors concentrations in this region which is characterized by high industrial emissions". I assume you mean something like "At PoValley sites the decrease was more pro-

nounced (-0.083 ppb/K/year) because of large reductions of precursor concentrations in this region which is characterized by high industrial emissions. Note that the average correlation between O3 and temperature in that cluster is the highest compared to the other regions".

* Lines 9-11 of page 19: Regarding the low correlations between O3 and T in the North cluster I would also mention the low temperature ranges observed there compared to the other clusters (Fig 11).

* Lines 12-16 of page 19. Discussion of the stronger trends of the O3-T relationship for the more polluted LT(t) clusters: "Our results are in line with a box-model study that tested the O3–temperature relationship under different NOx level scenarios (Coates et al., 2016). Coates et al. (2016) have shown that at high NOx conditions O3 increases more strongly with temperature, while the increase is less pronounced when moving to lower NOx conditions". As mentioned for the Po Valley S(t) cluster (see a couple of comments above), I don't see a clear relationship. These references are related to the strength of the O3-T relationship but not to the trend of that relationship. I assume that you mean that regional ozone production has mainly decreased at the most polluted locations, due to considerable reductions of precursor emissions there. Need to rewrite.

* One can remove the columns "standard deviation" in Tables 3 and 4. The p-values should suffice.

* In different parts of the text, the authors indicate that the S(t) clusters represent the "climatic conditions". I would add the word "regional" to clearly indicate that this is indeed a geographical classification that clearly reflects the regional climate conditions. This can be done in different parts of the text. Here I simply include an example for lines 5-6 of page 21: "Our approach captures several features of O3 variations, i.e. pollution level from the L(t)-clustering and influence of the climatic conditions from the S(t)-clustering". I would change the end of this sentence to "influence of the regional

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climate conditions from the S(t)-clustering".

* Lines 17-18 of page 21 (Conclusions): "peak O3 has been decreasing with the smallest rate at higher altitude sites especially in the western part of Europe due to the influence of background O3 imported from North America and East Asia". Are the evidences for this long-range-transport influence so clear? If not I would add the word "possibly" before "due to".

* Line 24 of page 21: "the sensitivity of O3 to temperature has weakened since 2000 with a rate of around 0.084 ppb/K/year". It should be considerably less than that because that value is only found for the 4 sites in the Po Valley cluster (see Tables 3 and 4).

* Lines 25-26 of page 21 (about the decreasing O3-T slope): "It was shown that differences in changes to this sensitivity across sites are mainly driven by regional meteorological conditions". I do not see any proof of this in the manuscript. It might well be related to varying rates of reductions of precursor emission across the different regions. I have a similar comment about this in the abstract so both of them can be addressed at the same time.

* The references Boleti et al. (2018a) and Boleti et al. (2018b) should be changed to Boleti et al. (2018) and Boleti et al. (2019), respectively. The second paper has been published and should be updated in the reference list.

* Figures S1 & S2: Can you please enlarge fonts. They are too small and very hard to read.

* Lines 2-3 of page 3 in Supplement: "objects have a low similarity with the rest objects" –> "objects have low similarity with the rest of the objects"

* Figure S3: Even when there are some explanations on the previous page, most readers are not very familiar with the concept of silhouette width. In the figure caption I would indicate (1) that each horizontal bar represents the silhouette width for a par-

ticular site in a given cluster and (2) that this parameter is clearly positive for most sites.

* Figure S5: I assume this is for W(t) instead of for S(t).

* Section S3 (Additional information on clusters): What is the use of this section if you hardly provide any comments e.g. about the MDA8 O3 clusters in the main manuscript? As indicated above, the main text and most of the supplement seem detached from each other.

* Lines 2-4 of page 10 in supplement: “In this section we present more detailed information about the clusters extracted from the daily mean and MDA8 O3 LT(t), S(t) and W(t)”. Please refer to Figures S9-S11 there.

* Caption of Figure S9: Need to remove “Map indicating the sites that belong in each cluster and average LT(t) in each cluster with the standard deviation of the sites that have SW>0”.

* Captions of Figures S10-S12: Need to use subscript for W in “SW” (silhouette width, defined in section S2).

* Lines 4-5 of page 14 in supplement: “The level off or small increase in the HighPoll stations can be attributed to the smaller rate of reduction of VOCs, which resulted to reduced titration of O3 by NO”. I agree with the reduced titration due to the decreases of NOx emissions, but do you have any evidence about the smaller rate of reduction of VOCs? If so one should provide a reference. A simpler explanation might be a change of chemical regimes, i.e. in the sensitivity of O3 production to NOx and VOCs.

* Line 6 of page 14 in supplement: Change “(Fig. S14” to “(Fig. S14)”. Then remove “respectively” because it is not needed there.

* Lines 10-11 of page 14 in supplement: “Here, the sites with negative SW that were not considered in the discussion of the trends are presented. In the LT(t)-clustering four sites with negative SW were identified (Fig. S15), in the S(t) 26 sites (Fig. S16)

and in the W(t) 24 sites". It is unclear whether you are talking about the clusters of daily O3 or MDA8 O3.

* Caption of Figure S15 is not complete. Need to indicate the meaning of the different lines/shading.

* Caption of Figure S16 is not complete either. For instance you don't show the "clusters average S(t)" as you claim because there is some spread in the figure. Probably mean +/- standard deviation?

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