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Interactive Comment

Interactive comment on "Origin of aerosol particles in the mid latitude and subtropical upper troposphere and lowermost stratosphere from cluster analysis of CARIBIC data" *by* M. Köppe et al.

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Reply to the reviewers comments and changes to the manuscript "Origin of Aerosol Particles in the Mid Latitude and Subtropical Upper Troposphere and Lowermost Stratosphere from Cluster Analysis of CARIBIC Data", ACPD 9, 13523–13567, 2009

Anonymous Referee #1

General comments: 1. ambiguous analysis, particle formation, winnowing of the data The referee is only partly correct, when he calls the presented analysis "ambiguous".

To our knowledge, for the free troposphere there has never been made such an attempt on a similar data set with so many parameters. Hence we were delighted, when we saw the cluster results for summer. However, even for summer there are of course data points e.g., from the boundary layer cluster, which could be attributed to the high clouds cluster, or vice versa. But this is not the important point. Cluster analysis does not always lead to clear results. The questions were, can the algorithm separate meaningful clusters? Yes, it mostly can. Can we interpret these clusters? Again, mostly yes. Hence, we felt the analysis to be valuable. Concerning the latter two points, the question about where particle formation takes place and the winnowing of the data, these are discussed in detail in the specific comments 2.5 and 2.1.

Specific comments: 2.1 P. 13528, L. 15-25, missing data and winnowing of the data: Yes, both referees are correct that the issue of winnowing the data needs more elaboration, particularly concerning what causes the missing values and how do they influence the analysis results. Hence, we changed the (now) third and fourth paragraph of Sec. 2.0 and hope we answered all the questions: "Unfortunately, in the data set there are many missing values for individual parameters, caused by instrument malfunctioning or calibration periods. Eventually the data set was reduced strongly by having to eliminate all data points from multivariate statistical analyses affected by missing values for individual parameters. For summer and winter seasons, defined below, only 26% and 38%, respectively, of the original data points could be used. The missing values occur mainly in the data of the three trace gases CH3CN, CH3COCH3, and NOy, i.e. data of two instruments. For these instruments between 40% and 70% of the missing values are caused by regular instrument calibration, which should not influence the results of the data analysis. Instrument malfunctioning is responsible for the rest of the missing values of the two instruments.

In order to be sure that the data reduction does not influence the following multivariate analysis, we compared histograms of the original data set and the reduced data set with respect to e.g., the geographical distribution, altitude, or the local time of day.

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Besides the geographical distribution of data points for summer, which shows a larger fraction of data points for the longitudes 20-50°E and 100-110°E, and a smaller fraction in the region 60-80°E compared to the original data set, all compared histograms look similar, which gives some confidence that the reduced data set is not strongly biased."

2.2 P. 13532, L. 4-24, the requirement of a normal distribution: Yes, both referees are correct. Cluster analysis does not necessarily require normal distributed data, but it might help the analysis. Concerning standardization, the text books about multivariate statistical analysis we refer to either "recommend" standardization before cluster analysis or state that "standardization must be carried out before cluster analysis". Hence we change the respective paragraph at the end of Sec. 3.1 to: "Before starting the multivariate analysis, two data processing steps were carried out. Although not necessarily required, normalization of the data might help data analysis (Leyer and Wesche, 2007). For that reason the original variables of the CARIBIC data subsets were examined for normal distribution by using histograms and significance tests (e.g., the Kolmogorov-Smirnov test). If the applied tests revealed a non-normal distribution, the corresponding original variable was transformed by applying either the logarithm, the square, or the radical function in order to bring it in a nearly normally distributed shape (Leyer and Wesche, 2007). After having evaluated each variable for normal distribution, the original variables were processed by centering and standardization. This step is recommended since measured variables often have highly different value ranges (Brosius, 2006; Backhaus et al., 2006; Leyer and Wesche, 2007). Centering and standardization leads to comparability of respective variables. For centering purposes the difference between the measured value and the arithmetic mean was calculated. Afterwards, within the standardization, this difference was divided by the standard deviation of the variable distribution. Accordingly, these processed variables (hereafter called z-parameters) have a mean of zero and a standard deviation of one."

2.3 P. 13533, L. 8-9, PCA factor extraction is only one possible method: Agreed. We changed the first sentence of the respective paragraph to: "PCA is one possible method to reduce the complexity of a data set by attributing as much as possible of the total variance of the original data to fewer newly defined variables (Brosius, 2006; Backhaus et al., 2006)."

2.4 P. 13541, L. 7-16, analysis of boundary cluster data points and biomass burning: A comparison between the spring and the late summer maxima in the biomass burning activity over Eastern Europe is not part of our analysis, since spring data are not included (we did not use the April flights and our summer season starts 2006 as well as 2007 late May). Clearly, a finer division into four seasons would help, but this would lead to poor statistics. Moreover, the detection of biomass burning plumes in the UT is a product of biomass burning activity and the frequency of efficient vertical transport mechanisms. Hence it would be interesting to see if the Eastern Europe biomass burning seasonal trend evolves into the UT, but one can not necessarily expect it.

2.5 P. 13542, L. 4-19, where does particle formation take place: The authors strongly believe that the observed particle formation takes place in the UT. Transport into the UT is mostly associated with clouds. However, nucleation mode particles do not survive this transport inside clouds, because either transport time is too long (e.g., in warm conveyor belts) or they are effectively scavenged by cloud droplets (e. g. in deep convective clouds). We try to indicate this position to the reader by changing the last paragraph of Sec. 4.2 to: "The nucleation mode particles, N4-12, can be regarded as an indicator for recent new particle formation events (cf. e.g. Hermann et al., 2003), as these particles have UT lifetimes of hours only (Williams et al., 2002). In summer, the stratosphere and the tropopause clusters show a similar range of N4-12 values of 260 - 1700 particles/cm3 STP, whereby the tropopause cluster distribution is shifted a little bit to lower concentrations (cf. averages in Tab. 3). Hence particle formation takes place in these regions. The high clouds cluster yields the broadest N4-12 distribution, indicative for clouds acting both as particle sink and as particle source (cf. Weigelt et al., 2009). The highest N4-12 values are found in the boundary layer cluster (1260 - 7530 particles/cm3 STP), a clear indication for particle formation in such air masses.

As transport into the UT is mostly associated with clouds, and nucleation mode particles do not survive this transport in clouds, because either transport time is too long (e.g., in warm conveyor belts) or they are effectively scavenged by cloud droplets (e.g. in deep convective clouds, Ekman et al, 2006), it is very likely that the boundary layer provides only the precursor gases but particle formation takes place in the UT. On the other hand, . . .”

Anonymous Referee #2

General comments: 3.1 uncertainties of cluster analysis: The present study was our first step in using multivariate data analysis tools, hence, we are not very experienced in this kind of analysis. This might be the reason, why we are not sure, if we understood the comment of the referee concerning the “uncertainty level for cluster division and cluster size”. Generally, there is no objectively best choice cluster analysis method, the results always depend on the data set, the subjectively chosen methods, and what the user regards as good and interpretable clusters. Hence we do not know how we should quantify an uncertainty. Concerning cluster number we believe, we clearly indicated that the choice was made according to how well we could interpret the results. Additionally, the “elbow criteria” has been used to verify our subjectively chosen cluster number. Concerning cluster size, clearly different cluster analysis methods would lead to different cluster sizes, however, how large this difference is, is hard to estimate without applying different methods. Hence, we will have to leave this comment open.

3.2 first data set over the Eurasian continent, size not larger than size of data sets from other campaigns: In this point we disagree with the referee. The measurement campaigns mentioned by the referee mainly took place at the borders of the Eurasian continent, either over Europe or south of India or east of East-Asia. Our data are the first measured really in the UT over the continent. Furthermore, please note that concerning valid aerosol data, not the reduced data file with only complete data points, we have more than 300 flight hours on the South-East Asia route. This aerosol data set is certainly larger than any data set obtained during a measurement campaign in the past. To emphasize, why we call our data sets the "first, large-scale seasonal data sets over the Eurasian continent", we added "(Eastern Europe, Central Asia, East Asia, and South-East Asia)" to the respective sentence in the summary.

3.3 statistically robust data set: Agreed. We removed the claim for the statistically sound data set and changed it to "large-scale seasonal" data set. The suggestion to combine the data from the South-East Asia route with those from the India route, which is currently done in another analysis on the CARIBIC aerosol data, is not feasible in this context, because these data were measured mainly with the old CARIBIC system, yielding no information on important trace gases like NOy, CH3COCH3, and CH3CN. Furthermore, the distance between the two average flight routes over Asia are more than 3000 km, which we believe is too much.

3.4 bias towards specific synoptic conditions due to commercial aircraft conditions: When looking at the flight tracks for individual flight sequences on the East-Asia route, one recognizes that the outward and return flights have nearly exactly the same track, at least for the Asian part of the flights. Furthermore, the majority of flights fall into a relative narrow flight corridor. Many flights avoid Russian territory and the Tibetan Plateau. Hence it seems that flight tracks are mostly determined by geography and not by specific synoptic conditions. On other flight routes, e.g., the North-America route the flight tracks spread over a much wider area. There, the synoptic conditions play a more important role in determining the actual flight track. Hence, for the data in this study, we assume no bias towards specific synoptic conditions. We inform the reader by adding the following two sentences to Sec. 2: "On this flight route, the actual flight tracks were mainly determined by geography and at least for the Asian part mostly fall into a relative narrow flight corridor (in contrast to e.g. the North-America flight route). Hence we assume no strong bias towards specific synoptic conditions, as it might be expected for commercial aircraft."

3.5 are results or clusters dominated by individual flights: We checked the clusters concerning dominating contributions by pairs of flights or flight sequences. For sum-

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mer, only the LMS cluster is dominated by two pairs of flights, both in early summer (end of May, mid of June), which contribute each about 33% of all data points in this cluster (clearly visible in Fig. 5). In winter, the boundary layer cluster is strongly dominated by the flight 166 (~55%), as already discussed in Sec. 4.2. Furthermore, the smaller FT cluster is dominated (~44%) by one pair of flights in March. All other individual contributions in both seasons are below 30%. To let the reader know these predominances, we added the following sentences to the cluster discussion in summer (Sec. 4.1) and winter (Sec. 4.2): Sec. 4.1: "The stratosphere cluster is the only summer cluster, which is strongly dominated by individual CARIBIC flight sequences, here namely 150-153 and 194-197, which contribute each about one third of all data points of this cluster. Consequently the cluster values should be regarded typical for early summer conditions. All other individual contributions of flight sequences to a summer cluster where below 30%." Sec. 4.2: "Interestingly, the majority of FT-2 data points were obtained in February/March (73%), while for FT-1 most of the data points belong to the period October-December (65%). The FT-2 cluster is dominated by one flight sequence, 224-227, which contributes about 45% of all data points to this cluster, while for the FT-1 cluster (as well as for the stratosphere and the tropopause cluster) the individual contribution of flight sequences is again below 30%.." and "This cluster and particular the data point cloud above 52° are strongly dominated by data from the CARIBIC flight 166 (Frankfurt - Guangzhou, October 19, 2006), which contributes about 55% of the cluster data points. This flight is characterized by strong pollution plumes and was already discussed by Slemr et al., 2009."

3.6 data set and local time of day: Good point. Yes, because of the fixed flight schedule of commercial aircraft and the mainly east-west flight route the data set is biased towards certain local times of day (both, the total data set including the missing values in the same way as the reduced data set without missing values). In winter, where this effect is stronger, 57% of the data points of the reduced data set fall into the six hours between 00:00 and 06:00. On the other hand there is a minimum between 15:00 and 21:00, with only 6% of the measurements. Data measured at mid-latitudes between

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40°N and 55°N are ∼80% night time measurements. The same holds true for data obtained below 23°N, which are ∼70% night time measurements. We notify the reader about this bias, by adding the following sentences to the end of Sec. 2. However, an interpretation on how this bias influences the data analysis is difficult, because such a large area, so many different geographical regions, and hence many different transport pathways are covered. “Interpreting the present data sets, one should be aware that because of the fixed flight schedule of commercial aircraft and the mainly east-west flight route the data sets are biased towards certain local times of day. In the winter data set, where this effect is stronger compared to the summer data set, 57% of the data points were measured in the six hours between 00:00 and 06:00 local time. On the other hand there is a minimum between 15:00 and 21:00 local time, representing only 6% of the data points. Measurements at mid-latitudes, between 40°N and 55°N, are about 80% night time measurements. The same holds true for data points below 23°N, which are about 70% night time measurements.“

3.7 a missing aircraft emission cluster: The reason, why we did not find an aircraft emission cluster is likely that aircraft emissions on our South-East Asia route are for a major part of the flight (besides Eastern Europe and South-East China) really at least a factor of two lower compared to the Caribbean route of CARIBIC (cf. e.g. the Global Aviation Emissions Inventory of the FAA, SAGE, http://www.faa.gov/about/office_org/headquarters_offices/aep/models/sage/). Even on the North Atlantic flight routes the clearly identifiable aircraft plume data were only 5-20%. Secondly, a minor reason, clear aircraft emission peaks in the aerosol number concentration are short, on the order of a few seconds only. Hence, due to the 10 s averaging in the meteo-merged files, clear and large signals might smear out. To inform the reader, we added the following sentences at the end of Sec. 4.0: “Further clusters, such as for aircraft emissions, possibly based on elevated NOy and particle number concentration values (Hermann et al., 2008), were not found with the described statistical analysis methods in the present data set. The most likely reason for not finding such a cluster is that the aircraft density on our specific South-East Asia route, although growing rapidly in the

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last few years, is still lower by at least a factor of two, compared to the highly dense North-Atlantic flight route (cf. e.g. the Global Aviation Emissions Inventory of the FAA, SAGE, Fig. 11, http://www.faa.gov/about/office_org/headquarters_offices/aep/models/sage/). But even there only 5-20% of the high particle number concentration peaks could be attributed to aircraft emission plumes (Hermann et al., 2008). Secondly, a minor reason, clear aircraft emission peaks in the aerosol number concentration are usually short, on the order of a few seconds only. Hence, due to the 10 s averaging in the meteo-merged files, clear and large aircraft signals might be diminished. These reasons do not exclude that on other flight routes an aircraft emission cluster might be found with the methodology described here."

3.8 no high cloud cluster in winter: The amount of clouds directly probed by the CARIBIC aircraft during summer and winter flights on the East-Asia route (indicated by non-zero values of H2O_cloud) is approximately the same, ∼13 %. But the high clouds cluster is not the cluster of data points inside clouds, but the cluster of data points which have been recently "influenced by clouds" (cf. Sec. 4.0). Actually, 72% of the data points in this cluster were measured outside clouds. Hence the difference between the summer and the winter clustering is probably caused by different cloud properties and cloud types, but not by cloud frequency. Apparently, high gas-phase and cloud water mixing ratios in summer helped to build-up a cloud cluster, while in winter these parameters were not so characteristic. We try to clarify this issue and changed the respective sentence in Sec. 4.2 to: "No high clouds cluster was found for the winter data set, which can not be attributed to the frequency of cloud encounters, which is approximately equal for the summer and winter data set. The difference between the two seasons is rather caused by different cloud types and cloud properties. In summer, a larger fraction of the clouds over the continent are deep convective clouds, which change the UT air more strongly than other cloud types do. In winter, the convective activity over the Eurasian continent is smaller (cf. ISCCP, http://isccp.giss.nasa.gov/products/browsed2.html)."

3.9 missing data and winnowing of the data: Please see answer to comment 2.1 of referee #1.

3.10 the requirement of a normal distribution: Please see answer to comment 2.2 of referee #1.

3.11 treatment of outliers: We do not agree with the referee that we did not explain how we defined outliers. In the manuscript there is nearly a whole page dealing with this issue. However, we willingly add the requested fraction of outliers to Sec. 3.1, namely, less than 0.4% for both seasonal data sets.

Specific comments: 4.1 L. 78-79, no global data set: Agreed. We changed "global" to "large-area".

4.2 L.117, interpolation of measurements with time resolution of more than 10 s: As only the mixing ratios for the two trace gases acetonitrile and acetone were interpolated, we changed the respective sentence to: "In order to have more complete data points, for this study the mixing ratios of the two trace gases acetone (CH3COCH3) and acetonitrile, which were obtained with a time resolution of 60 s, were linearly interpolated when the gap between following data points was smaller than 121 s. "

4.3 L. 146 and 161, "33" flights instead of "36": In table 2 there are in total 36 long-distance flights, where data from the CARIBIC container were available. However, in order to have clear seasonal results, we skipped the four April flights for the cluster analysis, which makes 32 flights, 15 in winter and 17 in summer. Hence the numbers given in the original text are correct.

4.4 L. 151-152, selection of flights by synoptic conditions: In our opinion, the attribution of flights to one of the two seasons is clearly described. Firstly, flights in June, July, and August were attributed to summer and flights in November, December, and January to winter. Even this attribution just by month was checked using the weather maps for the respective flight dates. As a division into four seasons was not reasonable (cf.

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next comment) and we wanted to improve the statistics by having more flights in a sub data set, we decided to expand the summer and winter seasons by adding flights from spring and autumn, if the synoptic weather conditions for the flight date allowed this. The April flights were not clearly attributable and hence were left out. We try to clarify the procedure by changing the respective paragraph to: "Previous studies have already shown that the concentrations of submicrometer aerosol particles undergo strong geographical and seasonal variations (e.g., Hermann et al., 2003). Therefore, a summer (15 flights) and a winter data set (17 flights) were extracted from the available data set. Because of the limited number of flights, each corresponding to only one specific meteorological situation in an overflown region, a finer division, e.g., into four seasons was statistically not reasonable. For the division not only the time of the year, but also the synoptic weather situation in the sampling area was considered. Firstly, flights in June, July, and August were assigned to boreal summer and those in December, January, and February to boreal winter. In order to get better statistics, we expand the summer and winter seasons by adding flights from spring and autumn, if the synoptic weather conditions for the flight date showed similar patterns like in summer or winter, respectively. Therefore we checked the synoptic meteorological situation indicated by the positions of controlling anticyclones, cyclones, and frontal zones of general flow patterns (Liljequist and Cehak, 1994; Gerstengarbe et al., 1999a; Malberg, 1997), pressure level maps (http://www.wetterzentrale.de; http://www.arl.noaa.gov), and model-derived vertical cross sections of the atmosphere (http://www.knmi.nl/samenw/campaign_ support/CARIBIC). The four April flights were not assigned, because the synoptic meteorological situation was ambiguous."

4.5 L. 152-153, division of data set: On the one hand side, we would like to have as much as possible seasonal information, i.e., a division of the flights into many sub data sets. On the other hand, although each flight provides hundreds of data points, it represents only one synoptic weather condition in an overflown region. Hence, we need a certain number of flights per seasonal sub data set to get good statistics. We think that two expanded seasons with approximately 16 flights each, are a good compromise between the two constrains. We clarified the “finer division” (s. previous comment).

4.6 L. 387-400, difference between boundary layer and the high clouds clusters in summer: We agree with the referee that a distinction between boundary layer and high clouds cluster points is difficult, because the high clouds of course transport boundary layer air into the UT. However, as Fig. 4 shows there are clear differences between these two clusters. The boundary layer cluster is the more polluted one with higher concentrations for acetonitrile, acetone, reactive odd nitrogen, and aerosol particles. Hence we prefer to discuss these two clusters separately. However, we indicate the similarities between the two clusters by adding the following sentences to Sec. 4.1: “Admittedly, there are data points in the boundary layer cluster, which could also be attributed to the high clouds cluster, or vice versa, as for instance deep convective clouds transport boundary layer air into the UT. Nevertheless, there are clear differences in trace gas and particle concentrations, which led to the separation of the two clusters.”

4.7 L. 423, pressure altitude and static pressure: The referee is correct that the pressure altitude and the static pressure are highly correlated, but some people prefer to think in pressure units some in altitude units when seeing vertically distributed data and hence we first kept both parameters. However this did not influence the data analysis, because in the course of the PCA (for the procedures 1, 2, and 3) this correlation is filtered, and either uncorrelated data (procedure 1) or none of both original parameters (procedure 2 and 3) went into the cluster analysis.

4.8 L. 467, biases in boundary layer cluster data: We are not sure if we understood the referee right. Of course, our data set is a limited one, which is representative only for the two summers 2006 and 2007, and the winters 2006/2007 and 2007/2008. Unfortunately, we have no influence, were our CARIBIC aircraft flies, this is entirely left to Lufthansa. Unfortunately Lufthansa decided 2008 to utilize the CARIBIC aircraft on another route, which closed the data set for the East Asia route. We indicate this limitation to the reader by adding the following sentence to the summary in Sec. 5:

“The data represent the two years period early summer 2006 to late winter 2008, and might look slightly differently for other years.” See also answer to comment 3.5 of this referee. Concerning the late spring biomass burning maximum over Eastern Europe please see answer to comment 2.4 of referee #1.

4.9 L. 477, comparison with concentration values from previous studies, same ranges used?: Unfortunately, we are afraid that we can not answer this point satisfactorily for the referee. In the past, particle number concentrations in the literature were mainly given as mean values or had to be estimated from figures. The authors are already happy that nowadays particle number concentrations for the UT/LMS are given in the same unit, i.e., particles/cm3 STP, which was not the case in the past. More recent papers (Singh et al., 2002 or Minikin et al., 2003) provide percentiles, but these can be 10%, 25%, 50%, 75%, or 90%-percentiles. Hence, without having the complete data set it is hard to calculate the same percentiles. Moreover, the particle counters used in the different studies had different lower detection diameters, which can lead to differences in the number concentration of tens of percent. Not to talk about the different, but mostly unknown or highly uncertain particle inlet transmission efficiencies. In our opinion, a difference between two aircraft-borne number concentration measurements of ultrafine aerosol particles (Dp $<$ 100 nm) of 50% or less can already called good agreement. There is much work to be done.

4.10 L. 516-517, comparison with INDOEX, ABC, ACE-Asia: The authors are aware of the three measurements projects, but we did not compare to INDOEX or ACE-Asia data, because the regions investigated during these campaigns are at least 2000 km away from the flight tracks of this study, both influenced strongly by pollution from the Asian continent. And for the measurements of ABC we are aware of only little data for the free troposphere (e.g. from the ABC-Pyramid Atmospheric Research Observatory in Nepal). Please remember that our data are restricted to 8-12 km altitude.

4.11 L. 516-517, difference in the flight routes in summer and winter: Yes that is true. For instance the more easterly route of the two flight routes over China was exclusively flown by the CARIBIC aircraft during the summer flight plan 2007 of Lufthansa. As this summer flight plan ended end of October 2007, there are two long distance flights (210-213) also in our winter data set. Most of the data are located within a corridor of ∼1500 km width, i.e. in the scale of synoptic-scale systems. Hence we think the different flight routes in summer and winter play a small role compared to the shift in atmospheric circulation associated with the seasons. We indicate this to the reader by adding the following sentences at the end of Sec. 2: "Most of the data points are located within a corridor of ∼1500 km width (Fig. 1), within the scale of synoptic-scale systems. Therefore existing differences in flight routes between summer and winter should play a minor role compared to the shift in atmospheric circulation associated with the seasons."

4.12 L. 563, differences between the two free troposphere clusters in winter: Yes, a deeper analysis of the differences between the two tropospheric clusters in winter could be interesting. But we have to note here that the presented data are a result of a master degree thesis, and hence time for analysis was limited. Nevertheless, about 50% of the data points in each of the two clusters were obtained from four flight sequences, which each contribute approximately the same individual fraction of data points to the summer and the winter cluster. Hence the different synoptic conditions between late autumn and late winter can only partly be responsible for the cluster differences. For the remaining flight sequences we checked the 250 hPa wind fields (http://www.knmi.nl/samenw/ campaign_support/CARIBIC), but this was not conclusive.

4.13 Fig. 4 and 7, different representation of Fig 4 and 7: We are not sure, what kind of presentation the referee meant exactly ("3D planes"?) . When we made figures 4 and 7, we already spent a long time in finding the best way to display the clusters. Using projections of the data point cloud on the coordinate planes (xy-, yz-, zx-planes) was one of them, but this would lead to one graph per cluster, which was not desired for the two figures. The figures should show all clusters in one graph, respectively, in order

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to get an impression on how well the cluster separation works. Hence we prefer the original presentation.

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