RC1

Review of
Statistical Analysis of Contrail to Cirrus Evolution during
the Contrail and Cirrus Experiments (CONCERT)

by Chauvigné et al.

The manuscript presents a new form of statistical analysis, the Principal Component Analysis (PCA), to investigate contrail to cirrus evolution based on two field campaigns. The observed ice clouds are divided in six clusters representative of different development stages of the contrails (primary wake, young contrail, contrail-cirrus and natural cirrus). Optical, chemical and microphysical properties of the clusters are then characterized to describe the ice-cloud properties during contrail to cirrus evolution.
Overall, the paper is very interesting and the topic is timely and suitable for ACP. Especially, the new approach to distiguish contrail and cirrus clouds seems to be promising. The manuscript is well structured and fluently to read. I found a number of minor points that I think needs claryfication before publishing the manuscript, they are listed below.

Nevertheles, my overall rating is major revisions because of two points emphasized here:
– I strongly recommend to include the RHI measurements of flight 19b during CONERT 1. They are published in Gayet et al. (2012), so the data are available – see comment 8).

– Some numbers in Table 2 needs to be checked, the mean and median IWC and, especially, the mean and median of Ntotal for CC are too high for natural cirrus – see comment 20).

We would like to thank the reviewer for his interesting and constructive suggestions. We have tried to follow every suggestion in order to improve the manuscript. Each reviewer's comments are addressed and the manuscript has been modified accordingly.

A significant change in the paper concerns the implementation of an automatic clustering method (k-mean method) to enhance the statistical significance of the contrail phase discrimination. Subsection "*3,2,2 Clustering analyses*" has been added and gives details of the method. After some tests we found that 16 clusters are necessary to classify the patterns revealed by the PCA analysis. To be in accordance with ATC observations, clusters 8 to 16 deduced from the k-mean method (Figure RC2.1.a.) were gathered into two clusters (A and B, Figure RC2.1.b.). Clusters 2 to 5 have also been merged to one single cluster.

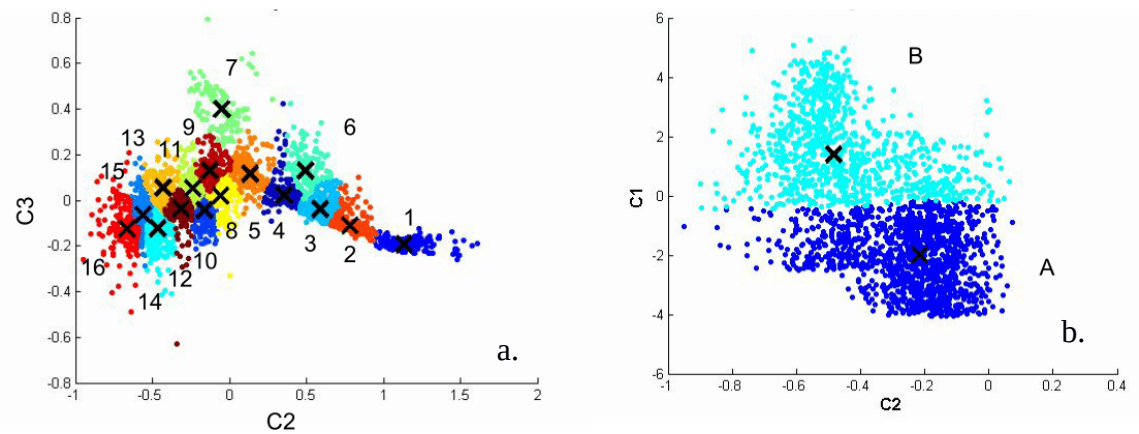

Figure RC1.1.: Clustering analyses according k-mean method. a) First step using 16 clusters and b) second step grouping clusters 8 to 16 to two clusters (A and B).

The following table summarizes the correspondence of the clusters defined by the k-mean methods and the cluster's definitions according to ATC information:

k-mean clusters	Cluster number	definition	name
1	0	Primary Wake	**PW**
6	1	Young Contrail 1	**YC1**
2, 3, 4 and 5	2	Young Contrail 2	**YC2**
A	3	Aged Contrail 1	**AC1**
7	4	Aged Contrail 2	**AC2**
B	5	Cirrus Cloud	**CC**

Table RC1.1: Cluster definitions according the k-mean method.

Comments:

1) Introduction: I like the very detailed introduction, but recommend to introduce more subsections (maybe even with titles), since now there are quite long paragraphs and the structure is not clearly visible.

The structure of the introduction has been changed and two subsections were added, namely:

1.1. Contrail formation and evolution

1.2. Optical and microphysical properties of contrail phases

Moreover, the description of contrail chemical properties has been shortened.
Modifications:
l.55:" *Several studies in the past have been dedicated to the evolution of concentrations of nitrogen oxide (NO) and sulphur dioxide (SO_2) and their oxidized forms (Kärcher and Voigt, 2006 ; Voigt et al., 2006 ; Schäuble et al., 2009 ; Jurkat et al., 2011).*"

2) Page 5, Line 194-195: *Particle size distributions and corresponding microphysical and optical integrated properties (IWC, Deff, N, and extinction) were derived from FSSP-300 measurements (Baumgardner et al., 1992).*
FSSP measurements does not include the larger ice particle and is thus know to be not suitable for calculations of at least IWC and Deff. The 2DC was also flown during the field campaign, so why not combining the two probes for the calculations? The missing size range between the two probes could be interpolated.

Optical and microphysical properties of contrail particles cannot be fully retrieved without the 2DC measurements. In the present study, these properties are retrieved with both instruments, FSSP and 2DC, to consider as much as possible the full-size range of the particle size distribution. The data are linearly interpolated in logarithm space between the two instrument ranges. The text has been clarified according to this remark.

Modifications:
l.191: "*Particle size distributions and corresponding microphysical and optical integrated properties (IWC, Deff, N, and extinction) were derived from both FSSP-300 and 2DC measurements.*"

3) Shattering of large ice crystals is negligible in contrails since the maximum size of the crystals is not large enough to cause this effect. I would mention that somewhere in the manuscript.

Indeed, the shattering of particles with size larger than typically 100µm on the probe inlet can affect the measurements. In addition, this effect can also explain the high number concentrations observed in natural cirrus (Table 3). It is now mentioned in the Manuscript.

Modifications:
l.235: "*These equations do not account for possible shattering of large ice crystals on the probe inlets. This effect is minimized in young contrails but can lead to an overestimation of small ice crystal concentration in contrail cirrus clouds.*"

l.647: "*Besides to interpolation between the FSSP-300 and the 2DC measurements, the assumed shape (spherical or aspherical), and shattering of large ice particles in cirrus and aged contrails can also have a significant effect on the measurement of optical and microphysical properties (Gayet et al., 2012).*"

4) Page 5, lines 220-221: *The bulk parameters were calculated assuming the surface-equivalent diameter relationships of Heymsfield (1972) and Locatelli and Hobbs, (1974).* Which bulk parameters do you mean?

The bulk parameters referred to the IWC, Deff, and extinction. It should be integrated or derived microphysical parameters. It was clarified in the text and, this sentence has been removed and the paragraph modified.

Modifications:
l.221: "*For spherical and non-spherical particles, the extinction coefficients are calculated from the following equation:*

$$Ext = \frac{\pi}{4}\sum_i \beta_{ext}^i N_i D_i^2 \qquad (1)$$

where β^i_{ext} is the extinction efficiency (values depend on spherical or aspherical particle characterization), D_i the mean diameter in channel i, and N_i the number concentration.

Different approaches are used to retrieve ice water content from spherical and non-spherical particles (Garret et al., 2003 ; Gayet et al., 2004 ; Gayet et al., 2012). For spherical particles (gPN > 0.85), IWC is computed from the following equation:

$$IWC_{spherical} = \frac{\pi}{6}\rho_{ice}\sum_i N_i D_i^3 \qquad (2)$$

with ρ_{ice} the bulk ice density (0.917 g cm^{-3})."

5) Page 6, line 234 - 238 : Calculation of IWC non-spherical : Is the validity of Equ. (6) ever checked by comparison to bulk IWC measurements?

The method used to derive IWC from spherical and non-spherical particles with equivalent diameter calculation, has been validated with different measurement techniques (FSSP-300 and polar Nephelometer) in the work of Gayet et al. (2012). The IWC derived from the particle size distribution is hindered by the uncertainties related to the size dependent enhancement factor for ice crystals in the inlet of total water instruments. There is a strong particle size dependence of the enhancement factor for bulk water instrument inlets in the small particle size range lower than 20 µm representative for young contrails. Further, there is little information on the particle shape effect on the enhancement factor. Finally, the inlet position of bulk phase instruments near the fuselage of

the aircraft may introduce additional ambiguities in the IWC measured near the aircraft fuselage. These uncertainties limit the quality of an assessment of the shape effect on the used mass dimension relationship using a comparison to the IWC measurements derived from bulk phase instruments.

6) Page 6, lines 254-256: *In addition, hygrometers using the Lyman-alpha technique (FISH, Zöger et al., 1999; Meyer et al., 2015), and frost point hygrometers (CR-2, Heller et al., 2017) were implemented on the Falcon during CONCERT-1 and 2.*
Please add the names of the hygrometers as indicated in blue.

The two hygrometers names have been added in the text.

Modifications:
l.248: "*(FISH, Zöger et al., 1999; Meyer et al., 2015), and frost point hygrometers (CR-2, Heller et al., 2017) were deployed on the Falcon during CONCERT-1 and 2.*"

7) Figure 1:
a) Caption: Time series at 1 s resolution for flights a) 19b (CONCERT 1) and b) 16b (CONCERT 2).
Please add the names of the campaigns as indicated in blue.

b) Plot of gPN: it would be helpful if a line at 0.85 would be drawn in the figure to better see if the particles are spherical or aspherical.

c) Plot of NO: a log scale might be better here, especially for Flight 19b from CONCERT 1.

All these suggestions have been considered. Figure 1 and its legend have been modified to improve the reading of the figure.

8) Flight 19b from CONCERT 1: Why are the RHI measurements of that flight not included here? They are published in Gayet et al. (2012), so the data are available.

RHI measurements performed during Flight 16b show typically values higher than (or close to) 100% when contrails are detected by the PN (extinction >0.1 km^{-1}). For Flight 19b, RHI values are always higher than 75% but values higher or equal to 100% are scarce. Moreover, contrail/cirrus events identified by the PN or chemical measurements do not seem to be correlated with RHI values. RHI measurements during Flight 19b and CONCERT 1 in general should be taken with caution. An additional bias in the temperature measurements of the Falcon may be responsible for an offset in the RHI measurements inside and outside of contrails during the CONCERT 1. This was not observed during CONCERT 2 where the temperature sensors have been extensively calibrated and RHI peaks near 100% are found in contrails and natural cirrus (Kaufmann et al., 2014). In addition, the descent of the primary wake within the wake vortices leads to an increase in temperature and thus an altitude dependent RHI profile within the contrails as observed by Gayet et al. (2012), Jeßberger et al. (2013) and Kaufmann et al. (2014). Thus, RHI measurements are shown for both campaigns but had to be analysed carefully taking into account the altitude of measurements and calibrations before flights.

Modifications:
RHI measurements for flight 19b (CONCERT 1) are added Figure 1 and Figure 3d.

I strongly recommend to include this data. It can be seen later in the paper that the number of RHI data from only flight 16b from CONCERT 2 is too low to apply the PCA analysis, see Figure 3, bottom right. Further, on page 8, lines 304-305 you write for flight 16 b: *However, no accurate ambient RHI data can retrieved for measurements in natural cirrus due to instrumental calibration problems.* but there are natural cirrus data available for 19b, CONCERT 1, yes?

We apologize for this possible misunderstanding. The PCA is solely based on light scattering measurements performed by the Polar Nephelometer. Other parameters such as RHI or NO concentration are used to validate or evaluate clusters/patterns revealed by the statistical analysis. On the principal component plots (Figure 3), "Natural cirrus" measured during CONCERT 1 are identified based on their scattering properties. Moreover, only one natural cirrus event was observed during flight 16b (none during flight 17) between 17:00 and 17:30. Unfortunately, no RHI measurements were performed during that time.

In addition, on page 6, line 252 you state the importance of RHI to characterize contrail ice crystals and on page 8, 3 rd paragraph, you describe how RHI influences the sphericity of ice crystals. So I think it is of importance not to leave out available RHI measurements!

RHI measurements during CONCERT 1 have been added to the manuscript. Instrumental issues reported in previous works (Kübbeler et al. (2011); Gayet et al. (2012); Jessberger et al. (2013) ; Schumann et al. (2013)) are also mentioned.

Modifications:
l.255: "*RHI measurements during flight 19b as well as instrument shortcomings are discussed in details in Kübbeler et al. (2011), Gayet et al. (2012), Jessberger et al. (2013) and Schumann et al. (2013).*"

9) Page 8, lines 294-297: *In a supersaturated environment of contrails, crystals grow by water vapor deposition and become increasingly aspherical with time. This is why spherical ice crystals prevail in very young contrails with an asymmetry coefficient around 0.85 with RHI above 100%.*

These sentences are a bit confusing. The reason that the ice crystals in young contrails are spherical under supersaturated conditions is that the time was too short to become aspherical, yes ? Maybe better: *In very young contrails, not enough time has passed so that despite RHI is above 100% spherical ice crystals with an asymmetry coefficient around 0.85 prevail.*

Indeed, the previous sentence was not that clear. We have modified the sentence following the reviewer suggestion.

Modifications:
l. 287: "*In a supersaturated environment, crystals grow by water vapour deposition and become increasingly aspherical with time. However, in very young contrails, spherical ice crystals with an asymmetry coefficient around 0.85 prevail.*"

10) Page 8, lines 305-309: *A good example of the evolution of gPN is the CRJ-2 contrail observed between 11:40 and 11:45 during flight 19b. The sequence illustrates the potential of the gPN measurement to characterize the evolution of contrail properties, with decreasing crystal sphericity documented by the decreasing asymmetry parameter from 0.88 to 0.79 (uncertainties around 0.04) after only 5 min and down to 0.77 after 20 min.*
Again, it would be very good to see the corresponding RHI measurements here.

RHI measurements for flight 19b are now added to Figure 1. We can see that despite the presence of a persistent contrail probably due to a supersaturated ambient air, RHI values during this period are too low. Thus, this example shows how the RHI measurements during CONCERT 1 cannot be considered to understand or detect contrail formation.

Modifications:
l. 294: "*However, it is important to note that the RHI measurements during the CRJ-2 chasing events do not show supersaturated conditions, whereas contrail seems persistent. Indeed, RHI measurements should be discussed carefully for this campaign due to calibration issues.*"

11) Page 8/9, last/first paragraph: Correlations between parameters are hard to recognize from Figure 1. Scatterplots for the main correlating parameters (gPN, RHI - from both flights, NO, extPN) would greatly improve the visualization of the discussed relations.

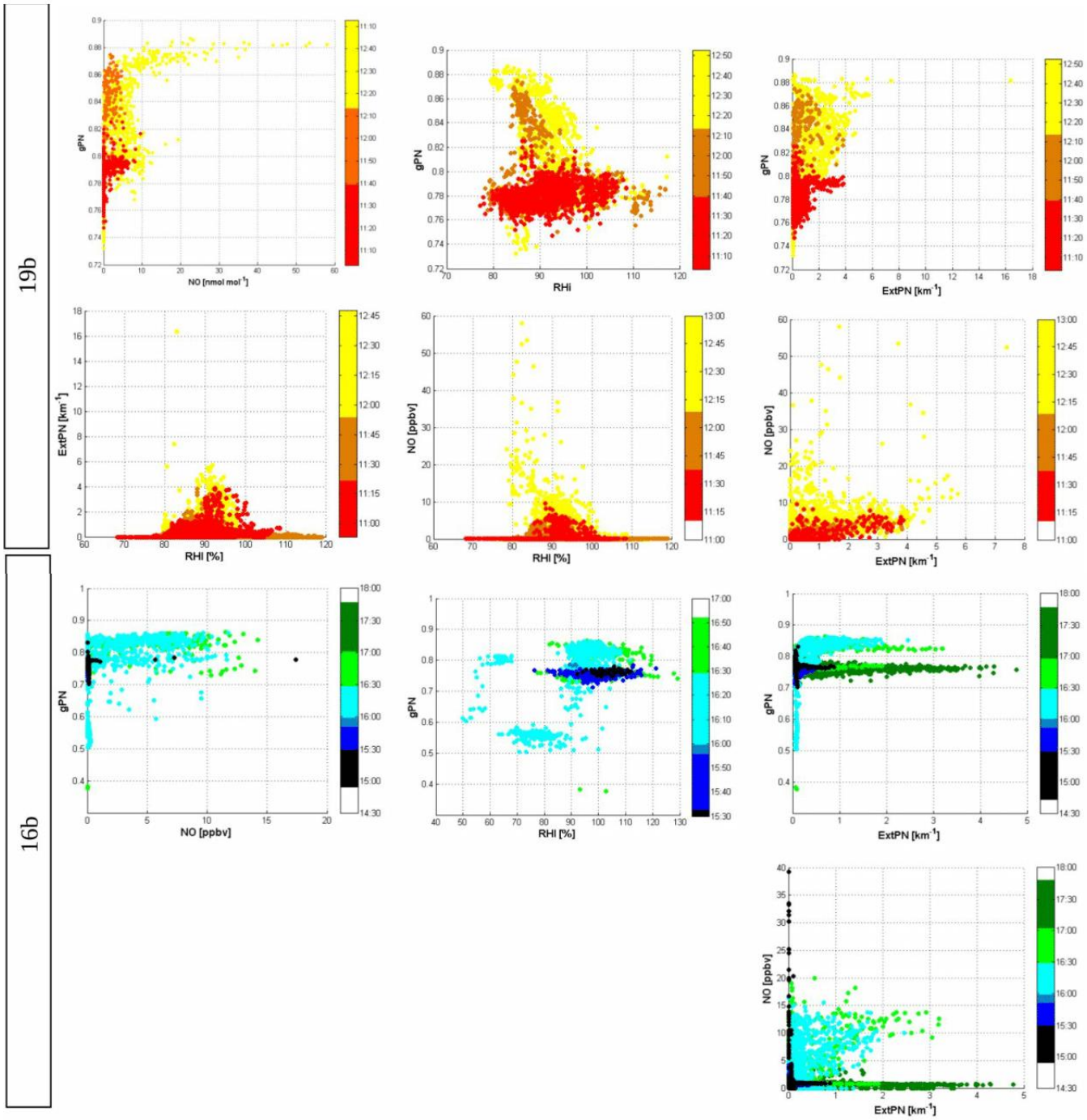

Figure RC1.2.: Correlation between optical and chemical properties during CONCERT 1 and 2 measurement campaign. Colours refers to ATC information for the different chasing and natural cirrus measurements.

The purpose of figure 1 is to illustrate the correlation/correspondence between the main contrail optical and chemical composition trends with ATC information. The only objective of this figure is to help the reader to understand how cloudy and clear sky conditions were determined and to show that parameters like the extinction, the asymmetry parameter and NO concentration can used to identify contrails and cirrus events.
To respond to the reviewer's comment, correlations between extinction, RHI, NO concentration and asymmetry parameter are shown on Figure RC1.2. for 19b and 16b flights. No clear correlations can be seen from these representations since each contrail stages are mixed together. However, NO concentrations and extinction coefficients seem to be linked for particular cases. For clarity, we decided not to show these correlations as it might misguide the reader.

12) Page 12, last paragraph: For a better understanding of this paragraph, I recommend
→ to make a table of the of the cluster numbers and the corresponding definitions (now listed at page 13, lines 453 – 458) and refer to the table at the beginning of the paragraph. In the present form, it is hard to follow the text without knowing the meaning of the numbers.
→ Further, it would be good to note the abbreviations of the numbers (0: PW, 1: YC1, 2: ...) in one panel of Figure 3, e.g. in 3a.

According to reviewer 2 suggestions the clusters have been redefined according Table RC1.1 and abbreviations of the numbers were also added.

Modifications:
The table of cluster definition is added (Table 1). Legends for all subplots has been added in Figure 6 in order to recall each cluster definition.

13) Page 12, line 420-421:
Figure 3a suggests an increase of Cj2 and a decrease of Cj1 with increasing aircraft size.
In Fig. 3a Cj2 vs. Cj3 is plotted, in the text you refer Cj2 vs. Cj1 – please correct.

Because the correlation is not as clear as mentioned in the text, this discussion has been removed from the text.

14) Definition of Clusters 3 (AC: Aged contrail) and 4 (ACC: aged contrail clean):
What is the difference between the two clusters? Does ,clean means low NO?
Please explain.

"Aged contrail" and "aged contrail clean" can be discriminated based on NO concentrations. "Clean" refers to low NO concentrations compared to measurements corresponding to AC conditions. AC and ACC have been replaced by AC1 and AC2 respectively without taking into account NO concentrations.

15) Figure 4: It would be helpful if you would include the circles from Figure 3 (a) in this plot.

Due to the modification of the clustering method, circles in Figure 3 have been removed. Every cluster's centre is now added on the new Figure 4.

16) Page 14, line 479: attribuate → attribute

The recommendation has been added into the all text.

17) Figure 5: I found it difficult to recognize the message of the panels of Figure 5. Here are some recommendation how this important figure can be improved:
a) in panel (a), a vertical line at gpN=0.85 would be helpful to distinguish between spherical and aspherical.
b) in panel (c), when using a logarithmic scale for the frequency the effects you discuss in the text will become better visible.
c) in panel (d), a logarithmic scale for NO and also the frequency would help to better see the the differences between the clusters.
d) in panel (e), a vertical line at RHI=100% would be good.
Further, in the text it is mentioned that the most frequent value of RHI is 95%. Shouldn‘t that be 100% ? And, the histogram is divided into small RHI intervals (2% ?), but the accuracy of the measurements is not better than 10-20%, I guess. I recommend to divide RHI in intervals corresponding to the accuracy and center them around 100%.
e) How many data points does each cluster contain ? This can be indicated in the legend.
f) The legend could be included in each panel – this would make it easier for the reader to assign the colors to the clusters when zooming the Figure on the screen.
Another idea could be to use more intuitive colors and sort the legend somehow into the stages of development, here a suggestion:
PW YC1 YC2 AC ACC CC

All these recommendations have been considered and included in Figure 6. RHI histogram has been modified to include RHI measurements of CONCERT 1.

18) Page 18, line 592:
Figure 6 shows mean volume particle size distributions (PSD) for all six clusters.
I see mean number PSDs – dN/dlogD19)

We apologize for this mistake, “volume” has been replaced by “number” into the text l.602.

19) Figure 6: The maximum sizes of PW and YC1 are already close to 200 µm, the maximum size of YC2 is close to that of ACC and CC. I would have expected smaller maximum sizes in the PW and YC categories, because ice crystals needs time to grow to larger sizes?
Further, the maximum size of CC is quite small – Voigt et al. (2017) show maximum sizes of natural cirrus PSDs up to 1000 µm or more ? See also comment 19 (b).

Indeed, no particle measurements have recorded up to 1000 µm during the two campaigns. Voigt et al. (2017) show “natural cirrus” and “contrail cirrus” with mean concentrations higher than 0.01 cm^{-3} for particles diameters around 100µm whereas concentrations do not exceed 0.01 cm^{-3} during CONCERT flights at this particle size range. Because instruments are different between the two campaigns, it can be explained by different shattering effects of largest particles, but also by air speed issues as explained in Febvre et al. (2009). In addition, young contrail particles exhibit diameters up to 200 µm. This effect can also be explained by the detection limit of the 2DC instruments which impacts the concentrations for very low signals (50% and 75% for concentrations of 5 and 0.5 cm-3 respectively, Gayet et al., 2002).
This discussion has been added to the text.

Modifications:
PSD measurements from Voigt et al. (2017) and Atlas et al. (2005) have been added to Figure 7.
l. 593: “*PSD measurements in natural cirrus and aged contrails differ significantly depending on the location of the study, ambient air conditions, measurement methods (instrument limitation (Gayet et al., 2002), and air speed (Febvre et al., 2009)). Previous studies show that a 3-hours old contrail cirrus with an effective diameter close to 20 µm (Voigt et al., 2017) and number*

concentration larger than 0.1cm^{-3} (Schumann et al., 2017) can be composed of ice crystals with sizes up to 100 µm (blue dashed line, contrail cirrus figure 7). This differs from the PSD of the natural cirrus presented by Voigt et al. (2017) (dashed black line), which has an order of magnitude lower particle number concentration. In natural cirrus at mid-latitudes, ice crystals with size up to 1600 µm were observed during the ML-CIRRUS campaign (dark dashed line Figure 7, Voigt et al., 2017)."

20) Table 2: (a) I suggest to sort the clusters like recommended under Point 16 f). A further suggestion is to sort Ntotal in two size intervals, namely <~30um and >~30um, since the grouping of the clusters change with size.
(b) The mean and median values of IWC does not fit to each other. For example, for PW / CC the means are 15.46/28.69 mg/m3, but the medians are 6.26/0.96 mg/m3, i.e. the mean of CC is almost twice the mean of PW, but the median of CC is much lower than that of PW ?
Please check all numbers.
(c) Mean/median of Ntotal for CC are 6.06/3.75 cm -3. This is too high for natural cirrus – from Voigt et al. (2017), I would expect something around 0.1 cm -3 or even lower. Is that an arithmetical error , shattering or could it be that contrails are accidentally attributed to CC ? Please clarify!

(a). Table 3 has been sorted according to the development of contrails as proposed by the reviewer in point 16.f).

(b). The new definition of the clusters does not allow to fit mean and median values. This observation may be due to the hypothesis used for PSD definition such as particle sphericity and the interpolation realized between 17 µm and 70 µm.

(c). As mentioned in the previous point, PSD of natural cirrus are significantly different according to measurement location and the different probes used. Here, the new clustering method shows lower number concentrations for the "natural cirrus".

Modifications:
l.623: "*Despite the large uncertainties associated to both instruments and the interpolation between 17 µm and 70 µm diameters, these results again show that each cluster can be connected to a specific contrail phase, and their properties can be compared to previous studies.*"

21) One last comment: could you discuss the possibility to use other/more parameters for the PCA? For example, could Ntotal be included in the PCA ? Or in case no Polar Nephelometer is on board, but PSD, IWC and NO is available, do you think the analysis would be possible?

The present paper demonstrates that the PCA method allows contrail classification from optical properties only. Indeed, additional parameter could improve the performance of the PCA method as PSD measurements, RHI values and NO concentrations. However, the additional parameter should be carefully selected in order to limit the bias introduced by the limitations of the probes and can vary from a measurement campaign to another.
It has been mentioned in the text.

Modifications:
l. 688: "*The additional use of microphysical and chemical measurements can be added to the PCA method in order to improve the selection of contrail phases. Different ranges of extinction or asymmetric coefficients could be also used for PCA analyses in this perspective. However,*

additional parameters should be carefully selected to limit the bias introduced by the limitations of the probes and the optimal selection may vary from one measurement campaign to another."