Responses to Referee#1 Darrel Baumgardner

of the manuscript ACP-2022-537: "Observations of microphysical properties and radiative effects of a contrail cirrus outbreak over the North Atlantic" by Wang et al.

The authors have improved somewhat upon their first effort and partially addressed my previous concerns; however, there remain a number of issues that must be rectified before I can recommend publication. These issues are all related to how the measurements are processed and interpreted from the CAS-POL and CIP, lack of a proper error analysis, and the failure to use the full capabilities of these instruments to distinguish contrail cirrus from cirrus.

We thank the reviewer Darrel Baumgardner for his helpful advices and constructive comments about our paper. The indicated issues have led to a revised version of our manuscript where we discussed about in situ measurements processing and uncertainty analysis, cirrus classification and the sensitivity on how the assumed crystal shapes, R_{eff} and IWC impact the simulated radiative forcing. To this end, we have written additional explanations and added a table.

In the following we number the referee comments (RC) and give replies (R) to each of them.

RC1: The criteria that is used to distinguish contrail cirrus from regular cirrus puzzles me, i.e. it appears that only the relative concentrations of NO and Nice, from the CAS and CIP) are used to discriminate the two types of cirrus. Previous studies, e.g. Järvinen et al. (2016) and Nichman et al (2016) has discussed how the CAS-POL polarization detection is sensitive to the shape of the small ice crystals, and the authors in the present paper also allude to the shape of ice crystals as sensitive to the type of cirrus, and yet neither the polarization ratio from the lidar or the CAS-POL is used to further separate the types of cirrus. Is this because this approach was tried but unsuccessful?

R1a) author's response

We thank the reviewer for pointing out this. The assessment of contrails and contrail cirrus particle shapes with cloud probes has been described and studied as part of the Contrail and Cirrus Experiment (CONCERT) in previous publications (Gayet et al., 2012 and Chauvigné et al., 2018). Here mainly young contrails were investigated and a clear trend in microphysical properties with aging was observed. The asymmetry parameter, derived with the polar nephelometer, shows a decrease with aging.

In this study, on one side we do not have measurements of the asymmetry parameter. On the other side, we know that particle shape depends on temperature and humidity as well as on the history of the ice crystals. Here mainly contrails and contrail cirrus of unknown origin (i.e. they could not be traced back to single aircraft) have been detected. The analysis of the shape in a similar way as in Järvinen et al. (2016) would require an enhanced effort, taking into account background atmospheric conditions and the atmospheric conditions along backward trajectories. It is out of the scope of the paper to do this analysis. Further the method to derive the shape for small particles would need a separate paper to discuss everything in detail. This is one of the reasons why we removed the descriptions and plots on asphericity in the first version of this paper.

RC2: Unless I didn't interpret what was written correctly, it appears that the CAS measurements below 3 μ m are not used in the analysis. I assume the thinking is that particles smaller than 3 μ m must be aerosol particles, not ice crystals. Whereas that might be a reasonable assumption, from contrail studies that I participated in during the early 1990s, we found that there were significant concentrations of contrail ice

crystals smaller than 3 μ m (Baumgardner et al., 1998). Similar studies by Kuhn et al. (1996), using the predecessor of the CAS-POL, also documented high concentrations of very small crystals. Then Kleine et al. (2018) also used a CAS-POL over the full size range to detect the smallest ice crystals. Hence, I want to see a reanalysis of the cloud passes using the full range of the CAS-POL since I hypothesize that the difference in contrail cirrus and cirrus will become much more distinct if you are only using number concentration. At the same time, I also hypothesize that the effective radius, Re, will also be much smaller in the contrail cirrus and provide a much more clear separation between regions with contrail cirrus and those without, particularly if you use the particle by particle data to identify fine scale entrainment and mixing.

R2a) author's response

It is correct that for this study only CAS-DPOL measurements starting at a particle size of 3 μ m were used. Measured particles smaller than 3 μ m were neglected. The focus of this study is the characterization of a contrail cirrus outbreak – the detection of individual, young contrails (seconds to minutes of age) was not in the scope of the flight strategy for this day due to many reasons. First of all, the operating aircraft HALO is not allowed to fly in the vortex of other aircrafts due to technical reasons. Second, the flight was operated in the North Atlantic flight corridor where HALO had to stay on defined flight paths and could not track individual aircraft pathways.

We agree that ice crystals smaller than 3 μ m are most relevant for the microphysical characterization of very young contrails (Voigt et al., 2010; Jeßberger et al, 2013; Kleine et al., 2018) but these young contrails were not detected during this flight.

Kleine et al. (2018) used CAS-DPOL data to study very young contrails with contrail ages smaller than 5 minutes. We removed data below 0.96 μ m due to technical issues. These small particles sizes can only be found in very young contrails.

Upon request of the reviewer, we have performed an addition analysis for R_{eff} to address your question through including particles sizes between 0.96 μ m and 3 μ m. After accounting for smaller particles, the mean R_{eff} for natural cirrus and contrail cirrus decreased by 2%. In contrast, the mean R_{eff} for contrails decreased by 8%. The difference of R_{eff} for contrail cirrus (contrails) and cirrus becomes a little more distinct.

In the manuscript, a short explanation for choosing a lower threshold of 3 μ m for the particle size was added. The authors would like to stay consistent with other studies of cirrus and contrail cirrus where a lower limit of the particle size was fixed to 3 μ m (Voigt et al., 2017; Righi et al., 2020). Thus, we would like to keep using measurements starting at 3 μ m also in this manuscript.

R2b) manuscript changes

L307-309: "Since the contrails detected during this flight are older, the lower threshold for the particle size was chosen to be 3 μ m to neglect any influence from aerosol particles in the size range below 3 μ m. This is consistent with other cirrus and contrail cirrus studies (Voigt at al., 2017; Righi et al., 2020)."

RC3: The use of Kleine et al, (2018) to define the uncertainty in size derivation as $\pm 16\%$ is valid for very small contrail crystals, but not for other crystals. As seen in the figure below, derived from Baumgardner et al., 2016, the uncertainty can be as much as $\pm 50\%$ due to asymmetries in shape. Given that the current study ignores shape as a parameter in defining cirrus types, this uncertainty is unimportant; however,

when comparing Re between contrail and non-contrail cirrus, it becomes important. In addition, the derivation of IWC will be very uncertain when you propagate this uncertainty in the calculation of IWC from the CAS size distribution. The derived IWC will exceed ±100%. Hence, since the N, Re and IWC are incorporated in the radiative transfer models, these uncertainties will need to be discussed in the model results.

R3a) author's response

We agree with the referee that uncertainties of the size derivation from the probe due to different shapes of the crystals translates into the IWC. This effect is known and we mention it now in the manuscript, thanks for pointing it out. However, R_{eff} rather than N and IWC from in situ data is used as input parameter for the radiative transfer calculations. The IWC for each measurement of R_{eff} corresponding to a vertically homogeneous ice cloud with given IOT from satellite observations is derived using Eq. (6) as the manuscript indicates.

We show below in Table A1 (Table 2 in the revised manuscript) that the R_{eff} and the shape of ice crystals have little effects on the radiative forcing in this case. In both cases, the reason is that the IOT of the pixels is kept constant according to the satellite observations. We finally decided not to update Fig. 7 as the lines from our sensitivity study overlap with the original ones. But we add a new paragraph with the related text at the end of Sect. 4.2 accordingly to address this source of uncertainty.

UTC/h	Shortwave RF / Wm ⁻²)				Longwave RF / Wm ⁻²				Net RF / Wm ⁻²			
	agg	-50%	+50%	ghm	agg	-50%	+50%	ghm	agg	-50%	+50%	ghm
6					54.1	53.9	54.0	55.1	54.1	53.9	54.0	55.1
7	-50.2	-50.7	-49.8	-47.5	90.4	90.0	90.2	91.3	40.2	39.3	40.4	43.8
8	-34.9	-35.3	-34.7	-33.7	50.4	50.2	50.3	50.8	15.5	14.8	15.6	17.0
9	-46.3	-46.6	-46.1	-44.8	49.0	48.9	48.9	49.5	2.7	2.3	2.8	4.7
10	-57.5	-57.7	-57.3	-56.1	40.2	40.2	40.1	40.6	-17.2	-17.5	-17.2	-15.5
11	-68.7	-68.9	-68.5	-67.6	40.4	40.5	40.3	40.8	-28.2	-28.4	-28.1	-26.8
12	-76.0	-76.2	-75.9	-74.5	40.5	40.6	40.4	40.9	-35.5	-35.6	-35.5	-33.7
13	-46.2	-46.2	-46.0	-45.1	31.0	31.0	30.9	31.2	-15.2	-15.2	-15.1	-13.8
14	-16.0	-16.0	-15.9	-15.4	15.0	15.1	15.0	15.1	-0.9	-0.9	-0.9	-0.3

Table A1: The sensitivity study on how changing the uncertainty of R_{eff} (± 50%) and assumed crystal shapes (aggregates agg and general habit mixture ghm according to Baum et al. (2014)) impact the resulting radiative forcing in Fig. 7.

R3b) manuscript changes

L126-128: "The uncertainty of the particle size measurements is $\pm 16\%$ (Kleine et al., 2018) for the lower size range and can be up to $\pm 50\%$ for R_{eff}, if the shape of the particles is not known. This translates into an error of up to $\pm 100\%$ for the IWC derived from scattering cloud probes (Baumgardner et al., 2017)".

L454: "... (called R_{eff,mean} in the following), ...".

L471-475: "To quantify how the estimated uncertainties in R_{eff} (and resulting IWC) as well as assumed crystal shapes influence the radiative forcing, we compute radiative forcing for aggregates (agg) with R_{eff} = $R_{eff,mean} \pm 50\%$ and perform a sensitivity study about ice crystal shape using the general habit mixture (ghm) also available from Baum et al. (2014). The simulated values are recorded in Table 2. The uncertainty

of RF due to R_{eff} has an average of 0.2 Wm⁻² in SW and 0.1 Wm⁻² in LW. In total the effect on the net RF is approx. 0.1 Wm⁻²..."

RC4: In section 4.1 (not 4.2 as is stated earlier in the manuscript), the radiative model uses an aggregate of ice crystals rather than a more reasonable mix of likely habits. If the average shape was aggregated crystals, where is the evidence from the CIP, which most certainly can identify such aggregates. Before I am willing to accept this very questionable simplification, I want to see a sensitivity study that show how changing the assumed crystal shapes impacts the resulting radiative fluxes. Likewise, I want to see how the estimated uncertainties in Re and IWC impact the flux calculations.

R4a) author's response

We thank the referee for this comment. We follow the suggestion and perform the sensitivity study on how the assumed crystal shapes impact the resulting radiative forcing. The parameterization named general habit mixture (ghm) from Baum et al (2014) is exploited to represent crystal shapes as a function of crystal size. The ghm with the R_{eff} of 25.2 μ m leads to a smaller absolute solar RF and a larger net RF than aggregates. However, the calculated IWC taking the crystal habits as ghm is larger than assuming as aggregates, and then results in a larger solar RF. These two parts cancel each other out in a way and generate a slightly larger net RF from ghm. In general, the impact of assumed crystal shapes on the simulated radiative forcing is small since we keep IOT constant. Additional changes in the text can be found below.

Notably, how the estimated uncertainties in R_{eff} and IWC influence the radiative forcing calculations are given in the answer section to comment RC3.

R4b) manuscript changes

L418: "A sensitivity study with respect to ice particle shape is conducted in Sect. 4.2."

L471-476: "To quantify how the estimated uncertainties in R_{eff} (and resulting IWC) as well as assumed crystal shapes influence the radiative forcing, we compute radiative forcing for aggregates (agg) with R_{eff} = $R_{eff,mean} \pm 50\%$ and perform a sensitivity study about ice crystal shape using the general habit mixture (ghm) also available from Baum et al. (2014). The simulated values are recorded in Table 2...Compared with aggregates, the ghm model has induced a larger net RF of 1.7 Wm⁻², with the shift in SW and LW of 1.2 Wm⁻² and 0.5 Wm⁻², respectively."

We hope to have addressed the important points raised by the reviewer in the revised version of the manuscript.

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Responses to editor

We thank the editor Farahnaz Khosrawi for her positive judgement on the manuscript and helpful technical corrections.

In the following we number the editor's comments (EC) and reply (R) to them individually.

Dear authors,

please find enclosed a referee report on the revised version of the manuscript. The referee has still some issues that should be considered/discussed before publication.

Additionally, I would like to ask you to consider the following technical corrections:

EC1: P2, L48-53: This sentence is quite long and difficult to follow. Please consider to shorten or split into two sentences.

R1a) author's response

Yes, we split it into two sentences.

R1b) manuscript changes

L48-53: "Due to various reasons, including the feedback of natural clouds, the radiative response to the presence of contrail cirrus, the uncertainty in upper tropospheric water budget (including initial contrail properties, contrail cirrus properties and relative humidity), contrail cirrus schemes (see Lee et al., 2021), and the challenges in measuring and separating contrail cirrus from natural cirrus, a best central estimate of the contrail cirrus RF remains challenging. It further limits projections of aviation climate impact and formulations of mitigation options other than carbon dioxide (CO2) emissions (Voigt et al., 2021)."

EC2: P3, L108-109: ...and broad and diurnal... -> one "and" too much? Better to use a comma?

R2a) author's response

This sentence means we exploit the information from high resolution (1) airborne measurements, (2) geostationary satellite observations to compute the diurnal cycle of RF in that region. Thus, the first "and" is replaced with "as well as".

R2b) manuscript changes

Updated the "...and broad and diurnal..." with "...as well as geostationary satellite observations with the high repetition rate..."

EC3: P3, L110: section should be written abbreviated as "Sect." except if written at the begin of the sentence, then it is "Section" (see ACP guidelines).

R3a) author's response

Updated all "section" in this paragraph with "Sect."

EC4: P5, L177: to 1% -> to be 1%.

R4a) author's response

Updated the text accordingly.

EC5: P6, Figure 1 caption: Full stop (last sentence) is missing.

R5a) author's response

The full stop "." has been added at the end of this sentence.

EC6: P6, L220: "usually missed" -> please rephrase.

R6a) author's response

Replaced "are usually missed" with "cannot be detected".

EC7: P6, L223: "under study" -> please rephrase.

R7a) author's response

Removed "under study" and replaced "the day" with "26 March 2014".

EC8: P6, L230: add a comma after "before".

R8a) author's response

Updated the text accordingly.

EC9: P6, L234: delete "the" before "Europe".

R9a) author's response

Updated the text accordingly.

EC10: P7, L238: perpendicularly -> perpendicular.

R10a) author's response

Perpendicularly is left here as we think that an adverb is required.

EC11: P9, L259: firstly -> first.

R11a) author's response

Replaced "firstly" with "first".

EC12: P10, L267: can be very probably identified -> can probably be identified

(or write "with high certainty")

R12a) author's response

Replaced "can be very probably identified" with "can probably be identified".

EC13: P11, Figure 4 caption: Add "the" -> over the North Atlantic and put the "N" before "< 3 mu m" and add "and" before "altitude".

R13a) author's response

Updated Figure 4 caption as "In situ measurements of HALO on 26 March 2014 over the North Atlantic region, including (a) ice number concentration N, (b) $R_{eff} > 1.5 \mu m$, (c) NO and NO background, (d) cirrus classification, (e) RHi, (f) flight latitude, and (g) altitude".