Reply to comments of Reviewer #2:

Thank you for the time and efforts you have spent on reviewing our manuscript; this is truly appreciated. Based on your comments (copied below) we reply with a point-by-point discussion of your concerns (italic, in blue). We also include a detailed description of how we have considered your suggestions in the revised manuscript version.

General remark: Because the calibration of radiation sensors in the laboratory and the required transfer of the calibration into the field by secondary standards cause additional uncertainties of the measured irradiances and the derived layer properties, we decided to use an in-flight calibration technique (instead of relying on the laboratory calibration and its uncertain transfer to the field) for the irradiance measurements. The in-flight calibration method was already successfully applied in previous field campaigns. It is based on radiative transfer simulations of the downward irradiance in clearly cloudless sky conditions at high altitudes. In this case the measurements are only slightly affected by the atmospheric layer above the sensor and the measurements can be adjusted directly to the simulations. This inflight calibration approach is then transferred to all radiation sensors installed in the aircraft and AIRTOSS. We have applied this calibration and concentrate on a specific measurement of the flight of 30 August 2013, which is more appropriate than the example discussed in the previous manuscript version. This specific case is characterized by a relatively high optical thickness of the cirrus layer taken into account that the vertical separation between the aircraft and AIRTOSS is 200 meters only. Another criterion for selecting this measurement case was that there was no additional cirrus above or below the layer enclosed by the aircraft and AIRTOSS. So we have chosen a case most suitable to derive optical layer properties of cirrus.

<u>Reviewer #2</u>: Experimental Setup: Figure 7: From grey shaded area in this plot (and Figure 5), the cirrus layer would extend beyond 1 km depth. The described experiment, where the AIRTOSS was extended a distance of _900 m (_185 m vertical), would not provide collocated radiation measurements above and below the cirrus layer.

Reply: You are correct, sorry for the confusion we caused here. This has not been described adequately in the manuscript. As the vertical difference between the two platforms is not more than about 200 m the investigated cloud layer, shown by the measurements in Fig. (7) and (8), represents a part of the complete cirrus layer. The revised sketch in Fig. (3) attempts a more clear explanation of the selection of the measurement case.



Fig. 3: Schematic sketch of measurement setup to measure collocated upward and downward irradiance at two altitudes.

Reviewer #2: In the region prior to that marked I and II, spaced by approximately 100 seconds of flight, was the AIRTOSS lowered even further? If it was not, then much of the supporting points given to support the use of AIRTOSS below and aircraft above (both of which were instrumented with radiation measurements), would describe a theoretical approach – not one that was enacted in practice. This should be made clear, and the title of the manuscript (": : :a feasibility study: : :" should be revised).

<u>**Reply:**</u> Thank you for the advice! The manuscript title has been changed to "Spectral Optical Properties of Cirrus from Collocated Measurements and Simulations".

We carefully screened the entire data set and selected a new measurement case (see Fig. 7) for the revised manuscript version. It represents a cirrus layer, that was directly between the two measurement platforms aircraft and AIRTOSS, thus not more than 200 m in vertical extent, which enables to measure above and below this cirrus layer.

Another reason is, that above and below the cirrus no additional cirrus layer was present during this part of the measurement flight.

Furthermore, this cirrus layer has a higher optical thickness compared to other measurement cases. Therefore, it shows even for the vertical extent of 200 m measureable and visible differences between the measured spectral irradiances.

Reviewer #2: In addition, ancillary support (microphysical data, other data?) for the region I as cloud free below cirrus, and region II as low-cloud below cirrus, is not given in the manuscript text. This would be an important point to address, given that an assumption of the analysis is that there is zero horizontal divergence of radiation – either within the cirrus layer or from high to low optical depth of the underlying (water) cloud. On this note, the assumption of zero horizontal divergence could be better established in the text. **Reply:** The new measurement example, as well as the complete measurement flight of 30 August, was influenced by a low – level cloud layer. This is supported by a video (recorded out of the cockpit) as well as synoptic forecast and MODIS data (see Fig. 4). That is why Fig. 7 shows one measurement example, with a low – level water cloud.



Fig. 4: (a) Composite satellite image of the cloud situation on 30 August 2013 at 9:45 UTC showing cirrus (white) above yellow colored lower water clouds (DeutscherWetterdienst / EUMETSAT).



Picture from out of the cockpit of the Learjet, showing the cloud situation during the flight. The low–level cloud layer is visible with the cirrus cloud above.

The assumption of neglecting horizontal components of radiative flux divergences in our approach was explicitly included in the revised manuscript version just after Equation 3, where we now state: "Eq. 3 implicitly assumes that there are no horizontal components of radiative flux divergence, only vertical flux divergences are considered to derive absorptivity."

<u>**Reviewer #2**</u>: Please also note that the level of the cirrus cloud was given different values in the manuscript (please check for consistency).

<u>Reply:</u> Okay, sorry for this error, which has been removed in the revised manuscript version.

Reviewer #2: Deriving Cloud layer optical properties from collocated measurements: Figure 8: As best I can gather, the downwelling irradiance above the cirrus layer came from a model, and not measurements. (In most spaces in text and captions, it is identified as a measurement, but in others, it is identified as model results). Besides the fact that it is unclear why a model has been used (given the discussion on a leveling platform in the manuscript), it must be considered that a mismatch between the measured and modeled downwelling irradiation above the cirrus layer would propagate into all of the derived quantities shown in the right hand plots. This mismatch would be an additional source of error that would propagate into the derived quantities because deriving these layer quantities is already subject to large systematic errors, given that the reflectivity and absorptivity are small values derived from differences in large irradiances. For example, the absorptivity results (subplots c and d) at wavelengths < 900 nm (i.e., conservative scattering wavelengths), show non-zero absorption when single scattering albedo at these wavelengths is essentially unity and true cloud absorption is expected to be zero. To achieve (expected) zero absorbtivity results at these spectral bands would require error bars that are 2-3X or more larger than the stated 5-6% uncertainty. Since the results of Figure 8 are (largely) derived only from measurements (the exception being the downwelling irradiance above cloud), I would ask for a comparison of measured and modeled downwelling irradiance above the cloud, and a percentage difference value between them.

Reply: There were some temporal problems with the active levelling of the optical inlet measuring the downward irradiance on the Learjet during the campaign. Therefore we have replaced the measured downward irradiances (Learjet) by respective simulations whenever the leveling platform did not work appropriately. That is well justified in case of measurements at high altitudes with no cirrus above. We have proven in numerous field campaigns that for such cases the horizontally levelled measurements are accurately described by simulations. This is not in contradiction with the general need to apply levelling platforms for measurements under more complicated cloud situations, which we avoided in the manuscript by carefully choosing a case with no cirrus above the high measurement altitude.

<u>Reviewer #2</u>: I would also argue that the upwelling irradiances below the cirrus base (subplot a) is larger than what would be expected for over-ocean, clear-sky measurements. What is the justification that low-level clouds were absent– what ancillary measurements did you use? And, how robust is the statement without support from lidar measurements, for example?

<u>Reply:</u> We have carefully screened all data available and on this basis have chosen a specific flight sequence, which is well appropriate to derive the cirrus layer properties. Finding such cases is not as easy as it appears from a first glance. As a result we came up with a new measurement case that we have investigated in detail. The new measurement example, as well as the complete measurement flight of 30 August, was influenced by a low-level cloud layer. The problem you have seen in the previous manuscript version does not appear anymore, mostly because the effects caused by the low-level liquid water cloud.



Fig.6: (a) Time series of downward (gray) and upward (light blue) irradiance $F(Wm^2 nm^{-1})$ measured on AIRTOSS at one wavelength (550 nm) from the flight of 30 August 2013. The thickened line periods mark the measuring points at straight flight legs. The red lines in (b) show the altitude of AIRTOSS (solid) and Learjet (dashed). The vertical dashed line marks the measurement example in Fig. 2.



Fig.7: (a) shows measured spectral downward and upward irradiance F from the aircraft above the cloud layer (solid lines) and AIRTOSS below the cloud layer (dotted lines) at the time, indicated by the vertical dashed line in Fig 1. Ftop is simulated. (b) shows spectral reflectivity (black), transmissivity (red), absorptivity (green), and cloud top albedo (gray) according to irradiance in (a). The vertical bars indicate the systematic errors due to measurement uncertainties.

<u>Reviewer #2</u>: Finally, given that the underlying surface albedo is ocean (in the absence of low-level cloud), I would ask that authors please remove all statements suggesting that variability in the cirrus reflectance is due to changing ground conditions given that ocean albedo is dark (low albedo) and relatively flat, spectrally.

<u>Reply:</u> Thank you for the advice! Please, see answers above relating the measurement example and the conditions below the investigated cirrus. We removed the statements criticized by the reviewer.

<u>Reviewer #2</u>: Comparing Measurements with Simulations: Please ensure that you make clear that the use of 1-D RT simulations will not capture horizontal radiation motion, potentially biasing your results (relative to "truth", as represented in a model by 3-D). Section 5.1: RT simulations – This section really needs more description to understand, with any certainty, the approach that has been applied. However, I believe this has been the general approach:

a) An (equivalent) particle diameter in conjunction with model single scattering properties and measured number size distribution, to compute bulk, volumetric layer properties. What were the assumptions to go from the non-spherical crystal shape to equivalent particle diameter?

Note also that the equations 7-9 should be revised to "dlogD" to correspond with the measurements in Figure 6.

<u>Reply:</u> The equivalent particle diameter (effective particle size) is defined as the ratio between the volume of the particle V(L) and the projected area A(L), where L is the maximum dimension of the nonspherical particle. A detailed explanation is given by Yang et al., 2005.

Yang, P., Wei, H. L., Huang, H. L., Baum, B. A., Hu, Y. X., Kattawar, G. W., Mishchenko, M. I., and Fu, Q.: Scattering and absorption property database for nonspherical ice particles in the near- through far-infrared spectral region, Appl. Opt., 44, 5512–5523, 2005.

<u>**Reviewer #2**</u>: b) The results from a) were used to model cloud optical properties, tau and reff. c) Derived tau and reff are then used to model upwelling and downwelling irradiance at cloud top and cloud bottom. d) Cloud layer properties are then derived from the irradiances computed in c). e) To investigate the cloud layer properties and cloud radiative forcing for different assumptions in crystal shape and habit mixture, a fixed number size distribution is combined with different shape assumptions.

Reply: Thank you for making this point. We have extended the explanation of our approach in the revised manuscript version. As you described correctly, the measured number size distribution is used to compute the volumetric optical properties. These bulk properties, e.g., extinction coefficient and phase function, serve as input for the radiative transfer model to derive the upward and downward irradiances at two altitudes, top and bottom of the cirrus. In turn the irradiances are used to calculate the layer properties transmissivity, reflectivity, and absorptivity, and the radiative forcing of the cirrus layer. By leaving the size distribution constant the ice crystal shape is varied to investigate its effect. In the revised manuscript version we have added a second scenario, which seems physically even more plausible. In this second approach we have kept the ice water content constant. For both scenarios we have investigated effects of changing ice crystal shape in detail. The description of the two approaches has been given in detail in the revised manuscript version.

Reviewer #2: One of my concerns with the results is focused on Figure 10 (albedo panel): None of the simulations, irrespective of crystal habit, reproduce the measured cloud top albedo. If I have understood the experimental process described above in points a-d, the failure to reproduce cloud top albedo calls into the interpretation of the layer properties and the cloud radiative forcing results.

<u>Reply:</u> The cloud top albedo is a measure of the optical properties of the complete atmospheric layer and the surface below. So, the properties of the low cloud influence the cloud top albedo as well and can only be reproduced poorly by the simulations due to a lack of knowledge about the lower cloud layer.

Reviewer #2: There are two, additional, key variables that are necessary to understand the results, that are I do not believe are provided in the manuscript: approximate optical thickness of the cirrus cloud, and solar zenith angle. For optically thin clouds, single scattering will dominate and assumptions in crystal shape will be emphasized (as opposed to an optically thicker cloud where multiple reflections smooths out some of the differences due to crystal shape/habit assumptions). In addition for low sun angles, multiple scattering will also be emphasized.

<u>Reply:</u> Your statements are correct, the degree of multiple scattering determines the magnitude of crystal shape effects. This is true in spectral regions with insignificant absorption, whereas in absorption bands multiple scattering amplifies shape effects. However, in this manuscript the focus is not to evaluate these dependencies by numerous sensitivity tests. Instead we take the measurements we have and try to come up with a consistent picture of the cirrus optical layer properties and related radiative effects for a specific measurement case, for which we have as many reliable measurements as possible. Then we investigate the effect of ice crystal shapes in Fig. 11 and for varying optical

thickness in Fig. 10, where different number size distributions are assumed, which mimics variations of cirrus optical thickness. We have not looked at the effects of different solar zenith angles, instead we have used the actual values during the measurements, which did not vary more than about 4 degrees.

<u>Reviewer #2</u>: Interpreting conclusions related to Changing Crystal Shape/Habit: I feel holding the number size distribution constant is an inadequate approach to quantifying differences in cloud layer properties. Understanding differences in cloud radiative forcing due to crystal shape and/or habit changes would require an approach that enforced constant IWC;

<u>Reply:</u> Thank you for the advice. As mentioned above we have added the scenario with a variable IWC into the revised manuscript. Please find the according layer properties and cirrus radiative forcing spectra in Figs. 10 (e) – (h). A respective discussion has been added to the text of the revised manuscript.



Fig. 13: Integrated values of cirrus radiative forcing when a low water cloud is present. The optical thickness (left panel), and the top height (right panel) of the water cloud are varied. The colors indicate a different cirrus optical thickness.

"The results obtained in this paper are valid for the respective cloud cases. To evaluate the low-level cloud effect on the cirrus the properties of the low water cloud, such as optical thickness and cloud top height, have to be investigated, too. Therefore, Fig. 13 (a) and (b) show values of integrated cirrus radiative forcings (wavelength range: 300–2300 nm) with varying water cloud optical thickness (a) and cloud top height (b). The cirrus is located between 6.7 km and 8.5 km altitude and consists of the mixture of shapes according to Baum et al. (2005). The color code represents the changing cirrus optical thickness.

In Fig.13 (a) the low–level cloud is located between 1 km and 1.25 km with an increasing optical thickness from 5 to 60. In general, the cooling of the cirrus decreases with increasing optical thickness of the low cloud resulting in an increasing influence of the low cloud on the upper lying cirrus. This is due to the reflected radiation by the low cloud being available to interact with the cirrus layer again. With higher water cloud optical thickness a saturating effect can be seen resulting in differences of 72% to 83%. Additionally, with increasing cirrus optical thickness the absolute difference of RF increases from 10Wm⁻² to 32Wm⁻².

In Fig.13 (b) the low water cloud has a constant optical thickness of 20, and a vertical thickness of 250m with an increasing cloud top height from 1.25 to 7.25 km. Similar to Fig.13 (a) the cooling of the cirrus decreases with increasing cloud top height of the low–level cloud. Here, the amount of the reflected radiation by the low cloud, available in the cirrus level, depends on the vertical extension of the atmosphere in between and its interaction with the transmitted (from cirrus) and reflected (from water cloud) radiation. The trend of RF is similar

to (a) with increasing cloud top height with a resulting in lower differences of 20% to 35% and absolute values of not more than 8Wm⁻². It is noticeable that the cloud optical thickness of the low cloud in comparison to the cloud top height has a significant effect on the radiative forcing of the above lying cirrus."

<u>Reviewer #2</u>: ... holding IWC constant was the approach also used by studies cited in this manuscript [Zhang et al., 1999] or to provide another boundary condition to results achieve by holding number size distribution constant [Wendisch et al., 2007]. To support the analysis results, plots of the spectral volumetric extinction coefficient, and single scattering albedo (as a function of crystal shape/habit, for perhaps 1-2 different particle diameters) should also be shown. The authors could also show the derived asymmetry parameter (first moment of the scattering phase function) as well.

<u>Reply:</u> We prefer not to add further graphs showing volumetric optical properties, this is beyond the focus of our manuscript. Instead we concentrate on the cirrus layer optical properties, resulting from measurement (microphysical) based simulations, and compare them with concurrent radiation measurements. We hope you will accept this decision.

The approach of assuming a constant ice water content, showing a more physical approach, was added to the simulations. In Fig. 11 different assumptions of ice crystal shape are investigated for the two approaches (I: constant number size distribution, II: constant IWC).



Fig. 11: Shown are spectral (a,e) transmissivity, (b,f) reflectivity, and (c,g) absorptivity of a cirrus layer between 6.7 and 8.5 km altitude. (e,h) are the radiative forcings at TOA, respectively. The simulations are based on a measured number size distribution assuming different ice particle shapes. The two panels indicate two conditions: constant number size distribution (Approach I) and constant ice water content (Approach II).