Observations of microphysical properties and radiative effects of a contrail cirrus outbreak over the North Atlantic

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Abstract. Contrail cirrus constitute the largest radiative forcing (RF) component to the total aviation effect on climate. However, the microphysical properties and radiative effects of contrail cirrus and natural cirrus clouds in the same meteorological conditions are still not completely resolved. Motivated by these uncertainties, we investigate an extended cirrus region perturbed by aviation in the North Atlantic Region (NAR) on 26 March 2014 during the Mid Latitude Cirrus (ML-CIRRUS) experiment. On that day, high air traffic density in the NAR combined with large scale cold and humid ambient conditions favouring the formation of a contrail cirrus outbreak situation. In addition, low coverage by low-level water clouds and the homogeneous oceanic albedo increase the sensitivity to retrieve cirrus properties and their radiative effect from satellite remote sensing. This allowed to extend current knowledge on contrail cirrus by combining airborne in situ, lidar and satellite observations.

In the synoptic context of a ridge cirrus, an extended thin ice cloud with many persistent contrails and contrail cirrus has been observed for many hours with the geostationary Meteosat Second Generation (MSG)/Spinning Enhanced Visible and InfraRed Imager (SEVIRI) from the early morning hours until dissipation after noon. Airborne lidar observations aboard the German High Altitude and LOng Range Research Aircraft (HALO) suggest that this cirrus has a significant anthropogenic contribution from aviation. A new method based on in situ measurements was used to distinguish between contrails, contrail cirrus and natural cirrus based on ice number and gas phase NO concentrations. Results show that contrail effective radii (R_{eff}) reach at most 11 μm while contrail cirrus R_{eff} can be as large as 51 μm. Contrail and contrail cirrus mean R_{eff} is 18% smaller than that of natural cirrus. ~~Results show that the effective radii (R_{eff}) of contrails and contrail cirrus are between 3 and 53 μm and are about 18% smaller than natural cirrus.~~ We find that a difference in R_{eff} between contrail cirrus and natural cirrus survives in this contrail cirrus outbreak event. As for radiative effects, a new method to estimate top-of-atmosphere instantaneous RF in the solar and thermal range is developed based on radiative transfer model simulations exploiting in situ and lidar measurements, satellite observations and ERA5 reanalysis data for both cirrus and cirrus-free regions. Broadband irradiances estimated from our simulations compare well with satellite observations from MSG, indicating that our method provides a good representation of the real atmosphere and can thus be used to determine RF of ice clouds. For a larger spatial area around the flight path, we find that the contrail cirrus outbreak is warming in the early morning and cooling during the day. The methods presented here and the results will be valuable for a future research to constrain uncertainties in the assessment of radiative impacts of contrail cirrus and natural cirrus and for the formulation and evaluation of contrail mitigation options.

1 Introduction

Aviation accounts for about 3.5% of global effective radiative forcing (ERF) from all human activities (Lee et al., 2021). Among the individual aviation contributions, contrail cirrus contributes to more than 50% of the total aviation ERF component (Lee et al., 2021). Contrail cirrus and natural cirrus are both high level clouds composed of ice crystals that form and evolve in ice supersaturated regions (ISSRs) (Minnis et al., 2004). Contrails form when the hot and humid jet engine exhaust at cruise

levels mixes with the cool ambient atmosphere, which at temperatures lower than the Schmidt-Appleman criterion (Schumann, 1996) can lead to a local liquid saturation of the plume. The emitted soot particles act as condensation nuclei to form liquid droplets that freeze subsequently in the young contrails (Bier et al., 2017; Kärcher and Voigt., 2017; Kärcher, 2018; Kleine et al., 2018). In ISSRs, persistent contrails may grow and spread out to form contrail-induced cirrus (Burkhardt and Kärcher, 2011; Schumann et al., 2017). By reflecting incident solar radiation and trapping upwelling radiation within the Earth's atmosphere, they result in an imbalance of radiation budget in both the shortwave (SW) solar and longwave (LW) terrestrial spectrum (Stuber et al., 2006). The net radiative forcing (RF) is positive (Rädel and Shine, 2008; Burkhardt et al., 2018; Gettelman et al., 2021) and RF due to contrail cirrus greatly exceeds that from linear contrails (Burkhardt and Kärcher, 2011; Voigt et al., 2011; Burkhardt et al., 2018). 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 limitsing projections of aviation climate impact and formulations of mitigation options other than carbon dioxide (CO_2) emissions (Voigt et al., 2021). Knowledge gaps still exist regarding the large variability in the contrail life cycles (Bier et al., 2017), and optical properties which then determine their radiative response to the climate system (Forster et al., 2007; Grewe et al., 2017).

Contrails may form in, overlap, merge, and interact with natural cirrus (Duda et al., 2001; Vázquez-Navarro et al., 2015; Sanz-Morère et al., 2021). Contrail cirrus primarily differs from natural cirrus by their larger ice number concentrations (N) (Heymsfield et al., 2010b; Voigt et al., 2010; Voigt et al., 2017; Sanz-Morère et al., 2020). Consequently, microphysical process rates, which control their life cycle, and radiative effects can be very different to those in natural cirrus and are dependent on soot number emissions (Bier et al., 2017). Furthermore, natural cirrus locally optically thickens by embedded contrails (Tesche et al., 2016; Quaas et al., 2021; Schumann et al., 2021a, b), but contrails can also cause a decrease in natural cloudiness (Burkhardt and Kärcher, 2011). These aspects are still subject of current research (e.g., Verma and Burkhardt, 2022). Aircraft and spaceborne measurements have provided detailed properties of contrail cirrus. Firstly, contrail cirrus can be detected and separated from natural cirrus to some extent in in situ measurements by combining ice crystal microphysical data with observations of aircraft emissions such as nitrogen oxides (NO_x) or aerosols (Voigt et al., 2017; Voigt et al., 2021; Bräuer et al., 2021a). Schumann et al. (2017) and Heymsfield et al (2010a) provide comprehensive overviews of contrail and contrail cirrus properties and extensive data sets exist on their microphysical properties (e.g. Petzold et al., 1997; Baumgardner and Gandrud, 1998; Jensen et al., 1998a, b; Voigt et al., 2010; Bräuer et al., 2021b), their particle shapes (Gayet et al., 2012; Järvinen et al., 2016; Sanz-Morère et al., 2020) and optical properties (Chauvigné et al., 2018), as well as the aviation influence on them (Jeßberger et al., 2013; Schumann and Graf, 2013; Marjani et al., 2022). Recent attempts used the reduced air traffic situation due to the COVID-19 pandemic to evaluate the aircraft impact on cirrus and climate (Gettleman et al., 2021; Li and Groß, 2021; Quaas et al., 2021; Schumann et al., 2021b; Meijer et al., 2022; Voigt et al., 2022). While the aircraft impact on clouds is confirmed by those studies, the magnitude of the reduced contrail cirrus forcing is variable and depends on the region, season and the method used to derive the impact. Some studies have taken the North Atlantic and North America with the largest air traffic density as target regions, and analysed the temporal evolution of contrail cirrus coverage, outgoing radiation, and properties (Duda et al., 2004; Atlas et al., 2006; Haywood et al., 2009; Graf and Schumann, 2012; Duda et al., 2013; Minnis et al., 2013; Schumann and Graf, 2013).

Early climate models estimated contrail cirrus RF through associating air traffic with regional cirrus coverage and assumed equal radiative efficiencies of contrails and contrail cirrus (Stordal et al., 2005; Rädel and Shine, 2008). Later, the global climate models represented contrail cirrus as a separate cloud class (Burkhardt and Kärcher, 2011; Bock and Burkhardt, 2016). In Chen et al. 2012, contrail cirrus is simply treated as a source for the ice crystal budget of the natural cirrus, mixing the microphysical properties of contrail cirrus and natural ice clouds. Hence, despite substantial progress in recent years, the

characterization of geometrical, optical, and microphysical properties of contrails and their evolution within natural cirrus fields as well as the calculation of the radiative impact are still subject to large uncertainties due to instrumental and model limitations and the large number of variables influencing the contrail life cycle (Chauvigné et al., 2018; Kaufmann et al., 2018; Rodríguez De León et al., 2018; Gierens et al., 2020).

In this study, we use in situ data measured during the Mid Latitude Cirrus (ML-CIRRUS) experiment (Voigt et al., 2017) from the German High Altitude and LOng Range Research Aircraft (HALO) and simultaneous remote sensing observations (Bugliaro et al., 2011; Vázquez-Navarro et al. 2013; Strandgren et al., 2017a) with high temporal resolution from the SEVIRI imager aboard the geostationary Meteosat Second Generation (MSG) satellite. In particular, we concentrate on one flight on 26 March 2014 over the North Atlantic Region (NAR) just off the coast of Ireland where most of the air traffic from Europe to the US and viceversa takes place. This situation enables us to investigate properties and radiative effects of contrail cirrus and ambient natural cirrus in an ideal contrail cirrus outbreak. In particular, we develop a new method to classify cirrus along the HALO flight track from in situ measurements based on enhanced NO aircraft gas emissions and ice number concentrations N into three representative classes: (1) contrails, (2) contrail cirrus and (3) natural cirrus where the effect of aircraft emissions is not directly observable. For these classes we evaluate microphysical ice crystal properties and relate them to relative humidity over ice (RHi). Satellite observations from MSG/SEVIRI are inspected visually to characterise the general cloud situation and confirm the presence of various contrails, in line with in situ measurements. In a new approach developed and for the first time presented in this paper, we determine the ice cloud RF that combines in situ and satellite observations with a radiative transfer model (RTM). To this end, we use reanalysis data from ERA5 (Hersbach et al. 2020). Since ERA5 does not simulate the effect of air traffic on clouds, we collect atmospheric profiles of water vapour, liquid and ice clouds from this reanalysis and combine them with the in situ and spaceborne observations to provide inputs to the RTM to compute reflected solar radiation (RSR) and outgoing longwave radiation (OLR). After checking the consistency of our radiative transfer calculations with the corresponding RSR and OLR measured with the Geostationary Earth Radiation Budget (GERB) instrumentation, we are in the position to compute instantaneous RF of this special combination of clouds in the SW and LW spectral range by excluding the ice cloud layer from the radiative transfer calculations, thus yielding a consistent ice-cloud-free irradiance. To be specific, we operate these calculations to an area encompassing the HALO flight path to compute the diurnal cycle of RF in that region, thus exploiting the information from high resolution airborne measurements as well as ~~and~~ geostationary~~broad and diurnal~~ satellite observations with the high repetition rate for a holistic view on the radiation budget from this ideal case in the NAR.

Detailed information about airborne and satellite datasets, as well as cirrus remote sensing techniques and the RTM are presented in Sect.~~section~~ 2. Microphysical properties of contrail cirrus and natural cirrus, consisting of cirrus classification, collocation of in situ and observed cirrus, and differences between contrail cirrus and natural cirrus, are summarized in Sect.~~section~~ 3. Radiative effects of contrail cirrus and natural cirrus, including the top-of-atmosphere (TOA) radiation estimation method, and the investigation of cirrus and RF spatial pattern are organized in Sect.~~section~~ 4. Finally, in the summary and conclusions in Sect.~~section~~ 5 we provide guidance about how to transfer our data evaluation techniques to future studies on radiative transfer calculations of contrails and contrail cirrus.

2 Data and approaches

2.1 Airborne measurements

During ML-CIRRUS, the German research aircraft HALO was equipped with a comprehensive suite of novel particle measurement sondes, and obtained a broad dataset of microphysical properties of contrail cirrus and natural cirrus for process studies and climatological analyses. Ice number concentrations N, R_{eff}, size distributions, ice or liquid water content (IWC and LWC) and extinction are derived from measurements of CAS-DPOL (Cloud and Aerosol Spectrometer with Detector for Polarization) for particles from 3 to 50 µm and CIP (Cloud Imaging Probe) for the size range of 15 and 960 µm (diameter as

the maximum dimension). CAS-DPOL measures the forward scattered light of particles when they pass through a laser beam (Baumgardner et al., 2011). The uncertainty of the particle size measurements is ±16% (Kleine et al., 2018) for the lower size range and can be up to ±50% for R_{eff}, if the shape of the particles is not known. This translates into an error of up to ±100% for the IWC derived from scattering cloud probes (Baumgardener et al., 2017). Using 64-element linear photodiode arrays, the

130   CIP acquires two-dimensional shadow images of particles (De Reus et al., 2009). The size resolution is 15 µm with the uncertainty decreasing considerably with diameter, reaching ±15 µm when particles are larger than 50 µm. N from CAS-DPOL, denoted by N_{CAS} in the following, and from CIP (N_{CIP}) are also combined to an overall N between 10^{-4} and 10^2 cm^{-3}. Validation of the measurements, have been performed taking atmospheric and cloud chamber measurements into account (Braga et al., 2017a, b).

135   As for ambient conditions, the AIMS (Atmospheric Ionization Mass Spectrometer, Jurkat et al., 2016, Kaufmann et al., 2016) was applied to measure the actual water vapor concentration from ambient air using a backward heated inlet. The range of detection is between 1 and 500 ppm with an overall accuracy from 7% to 10%. Static pressure and temperature, measured by the BAHAMAS (Basis HALO Measurement and Sensor System, Krautstrunk and Giez, 2012) with an accuracy of 0.3 hPa and 0.5 K, were used to convert water vapor concentration to RHi with an uncertainty of 10 % to 20 % (Kaufmann et al., 2018).

140   During the mission, nitrogen oxide (NO) and the sum of all reactive nitrogen species (NOy) were measured using the AENEAS instrument (AtmosphEric Nitrogen oxides mEAsuring System). This dual channel instrument is based on the well-established chemiluminescence technique combined with catalytic conversion of the NOy components. It has been regularly operated on HALO during several missions (e.g., Ziereis et al. 2022). A detector identical in construction has also been operated on a commercial aircraft as part of IAGOS-CARIBIC (Stratmann et al., 2016). The statistical detection limit is about 7 pmol/mol

145   for NO. The overall uncertainty depends on the ambient concentration of NO and ranges from about 8 to 6% for volume mixing ratios between about 1 and 0.5 nmol/mol. The time constant of the instrument for measurements in the upper troposphere is of the order of 1 s. The time offsets of NO have been shifted based on ice number.

Backscatter profiles of clouds and aerosol were acquired by the lidar system WALES (Water vapor Lidar Experiment in Space, Wirth et al., 2009) at the wavelengths of 532 and 1064 nm. In this study backscatter is used to extract information about the

150   cirrus cloud structures, such as cloud top height (CTH), geometrical depth and others. WALES also provides 2D measurements of the water vapor mixing ratio and aerosol particle linear depolarization ratio. The backscatter ratio and aerosol depolarization (perpendicular to parallel) (Biele et al., 2000; Esselborn et al., 2008) are used to create a cloud mask, which helps to discriminate for ice clouds. For these clouds the RHi is calculated from the measured water vapor mixing ratio and collocated model temperatures from the European Centre for Medium-Range Weather Forecasts (ECMWF). This instrument and method

155   have also been applied by Groß et al. (2014) and Urbanek et al. (2018), who found that the lidar measurements were accurate when compared with in situ data. The statistical error in the retrieval of water vapor by WALES is estimated to be about 5% (Kiemle et al., 2008) and the ECMWF temperatures induce an error of around 10-15% in the final RHi values (Groß et al., 2014).

2.2 Satellite remote sensing

160   The Spinning Enhanced Visible and InfraRed Imager (SEVIRI) is the primary instrument aboard the geostationary MSG satellites, which provides observations of the Earth disk every 15 min from 3 solar and 8 thermal channels with 3 km sampling distance at nadir, and one High Resolution Visible channel with 1 km spatial resolution (Schmetz et al., 2002). We use MSG-3 / Meteosat-10 observations for the study on 26 March 2014 with a temporal resolution of 15 min.

2.2.1 CiPS

165   CiPS (Cirrus Properties from SEVIRI) detects cirrus with their transparency information and retrieves the corresponding CTH, ice optical thickness (IOT), and ice water path (Strandgren et al., 2017a). It consists of four artificial neural networks trained

using SEVIRI thermal observations, CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) cloud products, and ECMWF ERA5 surface temperature and auxiliary data. CiPS has been especially developed for thin cirrus and validated against CALIPSO. CiPS detects 20%, 70% and 85% of the ice clouds with an IOT of 0.01, 0.1 and 0.2 respectively. For IOT between 0.35 and 1.8 CiPS has a mean absolute deviation smaller than 50%. This value increases for IOT between 0.07 and 0.35. For CTHs larger than approx. 8 km, CTH has an absolute percentage error of 10%, with underestimation for CTH > 10 km at 50° N and overestimation for CTH < 10 km at the same latitude. An example is shown in Fig. 1.

2.2.2 GERB and RRUMS

The GERB (Geostationary Earth Radiation Budget) instrument measures broadband solar and thermal components which are subsequently converted to outgoing and reflected fluxes considering the cloud properties and surface type detected by SEVIRI (Harries et al., 2005). GERB's sampling distance is larger than that of SEVIRI with a spatial resolution of 44.6 km × 39.3 km but the same image repeat cycle of 15 min. During the processing, the finer spatial resolution of the SEVIRI data is used to improve the original GERB resolution and results in GERB products for 3x3 SEVIRI pixels. In general, the GERB SW and LW fluxes are found to be 7.5% higher and 1.3% lower respectively, compared to products from the Clouds and the Earth's Radiant Energy System (CERES), whose data records are from polar orbiting satellites (Wielicki et al., 1996). The bias of CERES is estimated to be 1% and 0.5% for OLR and RSR, respectively.

Based on a linear regression and a neural network, an algorithm named RRUMS (Rapid Retrieval of Upwelling Irradiances from MSG/SEVIRI) was also developed, which estimates OLR and RSR at TOA from SEVIRI at pixel levels. RRUMS shows excellent agreement with OLR from CERES within 1% and a systematic overestimation of RSR from CERES or GERB of 5% to 10% in the worst cases under high viewing angles (Vázquez-Navarro et al., 2013).

2.3 Radiative transfer model

To calculate broadband solar and thermal irradiances at TOA for ice particles, the sophisticated radiative transfer package libRadtran is used (Mayer and Kylling, 2005; Emde et al., 2016). Water and ice clouds are represented in this model in detail and realistically. Optical properties of water droplets are computed using the Mie theory and tabulated as a function of wavelength and R_{eff}. Ice crystals are not spherical in shape and habits (Letu et al., 2016), and for this simulation the parameterisation of Baum et al. (2014) for ice crystal habits has been employed to define the conversion from optical to microphysical properties. In analogy to the MODIS products (Yang et al., 2018), we select rough aggregates for ice crystal shape (see Sect. 4.2 for a discussion about this choice). The selected one-dimensional radiative transfer solver is DISORT (Discrete Ordinate Radiative Transfer) 2.0 by Stamnes et al. (2000) with 16 streams. LibRadtran recommends the REPTRAN band parameterization with a spectral resolution of 15 cm^{-1} for spectral calculations (Buehler et al., 2010; Gasteiger et al., 2014).

We exploited positions of ice clouds from ML-CIRRUS measurements and CTHs estimated from WALES observations to collocate the clouds probed by HALO with the SEVIRI observed contrail cirrus and natural cirrus. We notice that satellite observations are functions of both time and space. Since the temporal frequency of in situ observations is 1 Hz, various HALO measurements are located inside each SEVIRI pixel. In general, time series of observations are produced with the original temporal resolution of the given instrument.

3 Microphysical properties of contrail cirrus and natural cirrus

In this section, we describe how airborne data from HALO and images from satellite remote sensing are combined to understand microphysical properties of contrail cirrus and natural cirrus, and the corresponding variation when contrails transit into contrail cirrus within the contrail life cycle.

3.1 General situation

In the night and the very early hours of 26 March 2014 a ridge cirrus cloud band built up North of Ireland down to the Southern tip of Portugal (Fig. 1). Close to Iceland as well as to the South-West of Ireland this cirrus cloud thickened (optical thickness larger than 1 at 10:45 UTC, Fig. 1d). While exactly off the coast of Ireland, in coincidence with the eastbound morning and westbound afternoon air traffic to and from the US, many linear structures can be seen in SEVIRI observations (Fig. 1, especially Fig. 1b where brightness temperature differences (BTD) between the SEVIRI channels at 10.8 and 12.0 µm are shown) and the ice cloud is thinner (optical thickness around 0.3 at 10:45 UTC). Please notice that Fig. 1 is enlarged in Fig. 2. Considering that the peak of eastbound morning air traffic is approx. at 3 UTC (Graf and Schumann, 2012), under favourable conditions with low temperature and high humidity contrails induced from these aircraft are expected to form and live for hours such that they can be identified in MSG observations in the morning of the same day. This cloud band evolves with time towards the South, and in correspondence of the Ireland coast the ice clouds dissipate around noon. In addition, this thin bluish/violet ice cloud band partly overlaps with a liquid water cloud field below (yellowish clouds in the false colour composite in Fig. 1a).

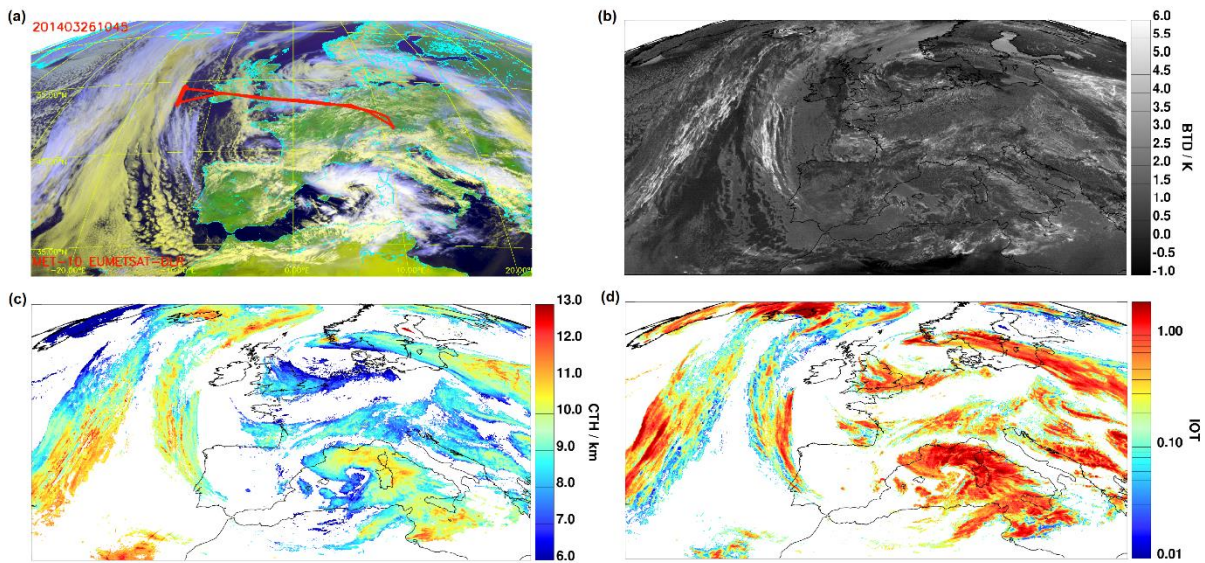



Figure 1: (a) The false color RGB image from MSG/SEVIRI overlapped with the HALO flight track on 26 March 2014 at 10:45 UTC showing Europe and the Eastern part of the North Atlantic Ocean, (b) corresponding 10.8 µm and 12.0 µm BTD (K) with overlaid ice clouds, (c) CTH and (d) IOT from CiPS.

Due to its approx. 3.5 km × 4.5 km spatial resolution, SEVIRI can only observe contrails that have already grown larger (and thicker) while young contrails ~~are usually missed~~cannot be detected (Mannstein et al. 2010). Nevertheless, the satellite observations indicate that because of these favourable meteorological conditions and the relatively high air traffic density in this region, contrails from various aircraft could form over a long time period (~12 h) that overlap with "the remnants" of previous contrails. Thus, air traffic in this area has a strong impact on cloudiness on 26 March 2014~~the day under study~~.

On 26 March 2014 the HALO aircraft started from Oberpfaffenhofen in Germany at approximately 05:30 UTC and probed the cirrus over NAR from around 08:00 to 11:30 UTC with a race track pattern between approx. 51.5°N and 54°N at a longitude of ca. -14°E (-13.6 to -14.4°E), see the flight track in Fig. 1a and also Voigt at al. (2017, Fig.4). In this area, HALO flew 3 lidar legs almost perpendicular to the NAR tracks (07:57 UTC - 08:35 UTC, south to north, 09:17 UTC - 09:30 UTC, north to south, and 10:21 UTC - 10:52 UTC, south to north), each followed by in situ legs at different altitudes.

Figure 2 presents the temporal variation of contrails and surrounding clouds with one HALO in situ/lidar leg at close time and air traffic data 2 to 3 hours before, from 08:30 (the first in situ leg) to 10:30 UTC (the third lidar leg). In addition, the area around the flight path is investigated in Sect. 4.2 in more detail but that serves here as orientation to easily capture the temporal evolution of the ice clouds. The wind speed in the east-west direction fluctuates around 0 while winds in the north-south

orientation drive the ridge cloud to move southwards (blue arrows in Fig. 2 indicate wind direction at 225 hPa from ERA5). The aircraft flight to and from ~~the~~ Europe. In the BTD images – where SEVIRI brightness temperatures at 10.8 µm and 12.0 µm are subtracted from each other - the black areas are caused by the low-level clouds, while the bright pixels correspond to

240   thin cirrus. Small ice crystals, for instance in contrails, correspond to the largest BTDs. From 08:30 UTC, many thin lines (contrails) are seen in the false colour RGBs in the first column in Fig. 2, where also the HRV channel was used. They run parallel to most air traffic routes (blue diamonds in the second column, S1) and are intersected perpendicularly by the HALO route. Some of the contrails are also visible in the BTD pictures (the second column in Fig. 2), that have a lower spatial resolution than the HRV channel, thus indicating the presence of small ice crystals. These contrails form a thin cirrus layer,

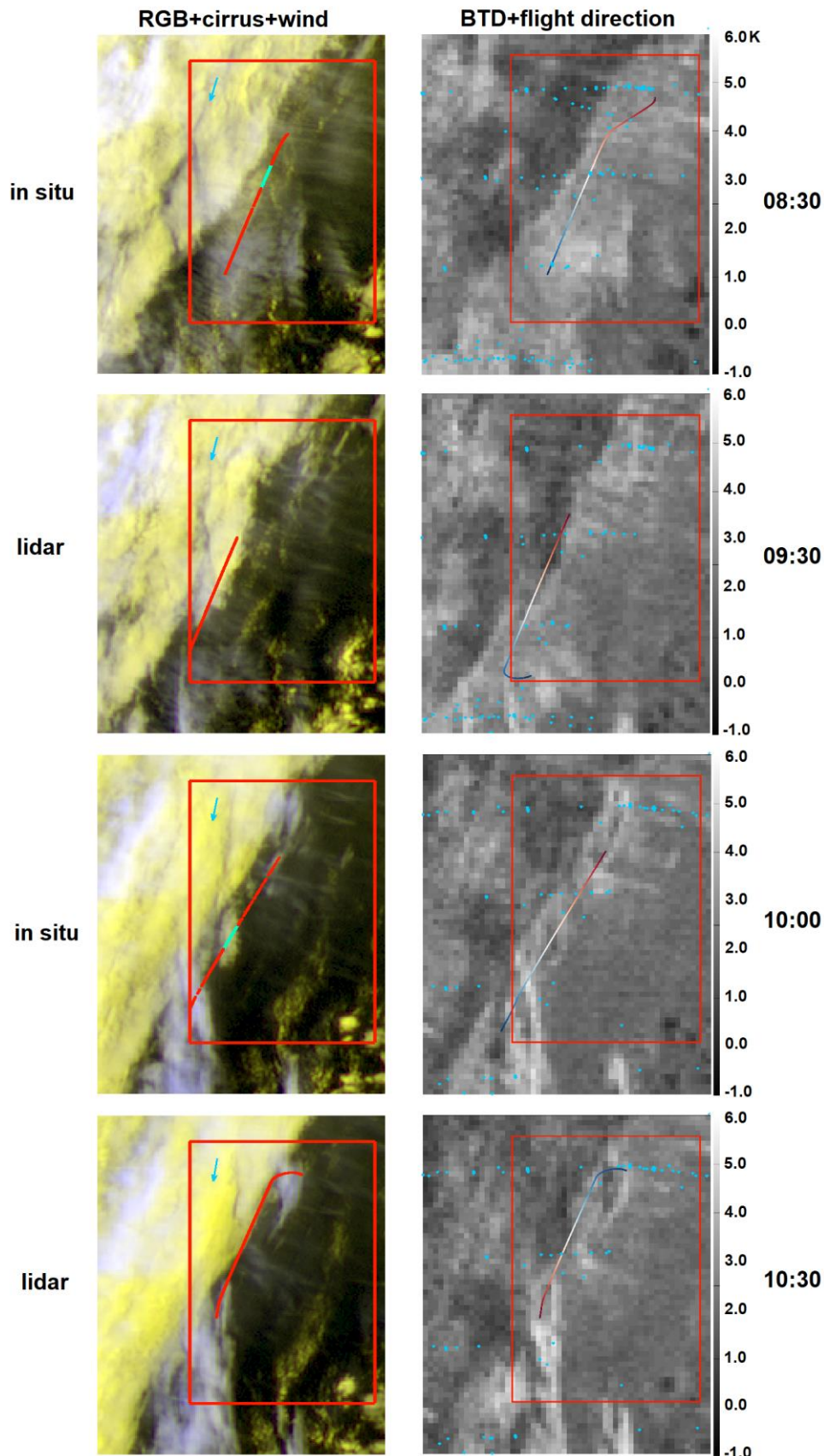
245   potentially of anthropogenic origin but also with natural ice clouds (see discussion of Fig. 3 below). In the South, the ridge cirrus is thicker and no contrails can be observed. At 09:30 UTC, the flight area is dominated by contrails. With time, from 08:30 to 10:30 UTC the typical contrail lines become always fainter and less in number. Thus, contrails begin either to dissipate or to lose completely their linear shape due to wind shear such that they turn out to be undistinguishable. However, contrails can be still observed in the north-eastern part of the satellite images at later times.

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Figure 2: Time series of contrail cirrus and surrounding clouds from MSG/SEVIRI observations over the NAR corridor on 26 March 2014. The first column: RGB-composite with overlaid cirrus, low-level liquid clouds pixels and in situ/lidar HALO leg at close time. The red and green line of the HALO flight track represent contrail cirrus and natural cirrus, respectively. The blue arrow indicates the wind direction almost perpendicular to the line shaped structures of the contrail cirrus. The second column: 10.8 μm and 12.0 μm BTD (K) with overlaid cirrus pixels. Blue points show air traffic dataset interpolated to MSG grid from M3 and NATS. The color of the HALO flight track indicates the flight direction. HALO flies from red to blue part. Top to bottom: 08:30, 09:30, 10:00 and 10:30 UTC. The red area is investigated in Sect. 4.2.

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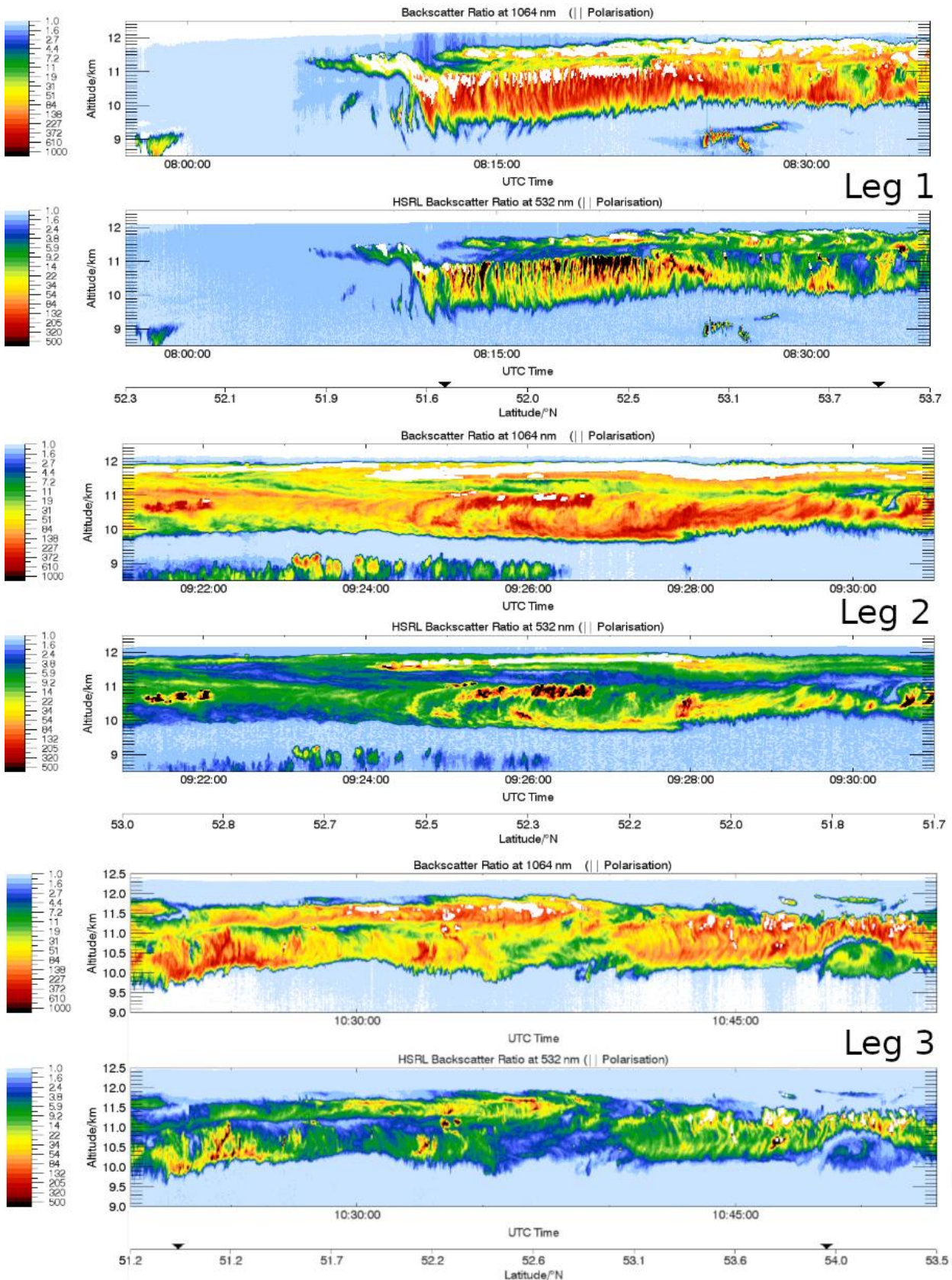


Figure 3: The three panels show the three lidar legs with backscatter ratios at 1064 nm and 532 nm.

Figure 3 presents the backscatter ratio at 532 and 1064 nm from the WALES lidar measurements in that sequences. The lidar data show that the geometrical thickness of high-level clouds reduces from ~2.0 km at 08:30 UTC to ~1.5 km at around 11:00 UTC. The temporal evolution of CTH from WALES shows that the cloud firstly reached up to approximately 12 km and slowly descended to slightly above 11.5 km, with backscatter values becoming smaller with time, in line with the passive

observations that indicate dissipation of the cloud during the day. For the investigation of contrails from in situ measurements we concentrate on the time period between 08:30 and 11:30 UTC, but we extend this time to cover the period from 6:00 UTC to 14:00 UTC in Sect. 4.2.

Considering the three WALES legs in more detail (Fig. 3), in leg 1 between 08:10 and 08:25 UTC there is a series of connected fall streaks with high backscatter at a height of 11 km below a thin ice cloud layer top at 12 km. These structures resemble those in the large eddy simulations by Unterstrasser et al. (2017a, b) where they considered contrails within natural cirrus. Thus, these bright spots with their fall streaks can ~~be very~~ probably be identified as contrails, with many small ice particles causing high backscatter and with larger ice crystals sedimenting down. For leg 1, these contrails make up the majority of the cloud in this temporal interval. The presence of developed fall streaks is a hint that these contrails are aged (> 30 min). Unfortunately, the high WALES backscatter inhibits the determination of RHi (Fig. S1) for these bright spots, but the upper level of the clouds, just below the potential contrails, show occasional ice supersaturation, with RHi above 100%.

Lidar leg 2, much shorter than the other two, also shows the presence of some spots with elevated backscatter ratios, although less numerous, at an altitude of 11 km at 09:26 UTC. Unlike leg 1, many high backscatter spots do not show extended fall streaks, suggesting the presence of younger contrails in addition to older ones. Furthermore, the top ice layer above 11.5 km also contains some very high backscatter areas. Finally, also above 9 km some clouds are visible that resemble contrails (or a dissipating cirrus). Thus, in this leg the contrail-like structures do not fill the entire cloud as in leg 1, but there are indications of young and older contrails, in addition to natural cirrus. The last lidar leg 3 took place 1 h later and also shows various bright spots at different levels, from 10 to almost 12 km, and elongated vertical structures that remind of those in leg 1. The cloud as a whole is slightly lower than that in leg 1 with ragged edges especially at its lower border, suggesting that the cloud is sublimating and thinning out. This is confirmed by the RHi observations of WALES for leg 3 in Fig. S2, where subsaturation is indicated especially at the lower and upper edges, while around 10.5 km at selected locations RHi reaches saturation. These WALES measurements are indicative for the cloud probed between 08:00 and 11:30 UTC but cannot directly be compared to in situ observations taken at different times in between.

3.2 Properties of contrails, contrail cirrus and natural cirrus

3.2.1 Identification of contrails and contrail cirrus and natural cirrus

In this section, we improve a method already used in Voigt et al. (2010) to identify where aircraft emissions are fresh, older or not detectable by using in situ measurements. These three situations are assigned to contrails, contrail cirrus and unperturbed cirrus, also called natural cirrus. The goal is to investigate microphysical and optical properties of these clouds. Figure 4 shows the full sequence of the airborne in situ measurements of N (N_{CAS}, N_{CIP}), R_{eff}, NO, cirrus (identified using the method in this Sect.), RHi, latitude and altitude as a function of time between 08:30 and 11:30 UTC. N for both instruments, CAS and CIP (N_{CAS} and N_{CIP}) and R_{eff} (Fig. 4a, b) indicate the occurrence of cirrus at the flight levels between 10 km and 13 km (Fig. 4g). According to the dilution equation by Schumann et al. (2017) and assuming 1000 particles/cm^3 for 1s-old contrail, many contrails probed here are more than 1 h old. Furthermore, some contrails are also visible in the low-resolution MSG/SEVIRI satellite images (BTDs in Fig. 2) and therefore at least 1-2 h old (Vázquez-Navarro et al., 2015). Figure 4c thus shows variable NO concentrations and various peaks. Since other sources of NO are unlikely in this situation (no thunderstorms in the previous 12 h, no wildfires), we assume that the NO concentration increases correspond to aviation exhausts, with different peak heights caused by various factors such as contrail age, dilution and aircraft type (Voigt et al., 2010; Jurkat et al, 2011; Jeßberger et al., 2013; Schumann et al., 2013). In fact, after the emission, NO is mixed with the surrounding air and dilutes with increasing plume age such that its measured mixing ratio can be used as a rough indication for contrail age. Also, for contrails initial ice particle concentrations N are high due to the high number of soot particles emitted by the engine (Schlager et al., 1997; Kleine et al., 2018; Bräuer et al., 2021a) and N decreases due to dilution (Schumann et al., 1998) and further processes (Bier et al., 2017; Unterstrasser et al., 2017a). High NO emissions without coincident ice crystal observations correspond to situations

where the Schmidt-Appleman criterion is not satisfied and no contrail is formed. This study, similarly to Voigt et al. (2010), uses N and NO to evaluate contrail evolution. Since the contrails detected during this flight are older, the lower threshold for the particle size was chosen to be 3 µm to neglect any influence from aerosol particles in the size range below 3 µm. This is consistent with other cirrus and contrail cirrus studies (Voigt at al., 2017; Righi et al., 2020).

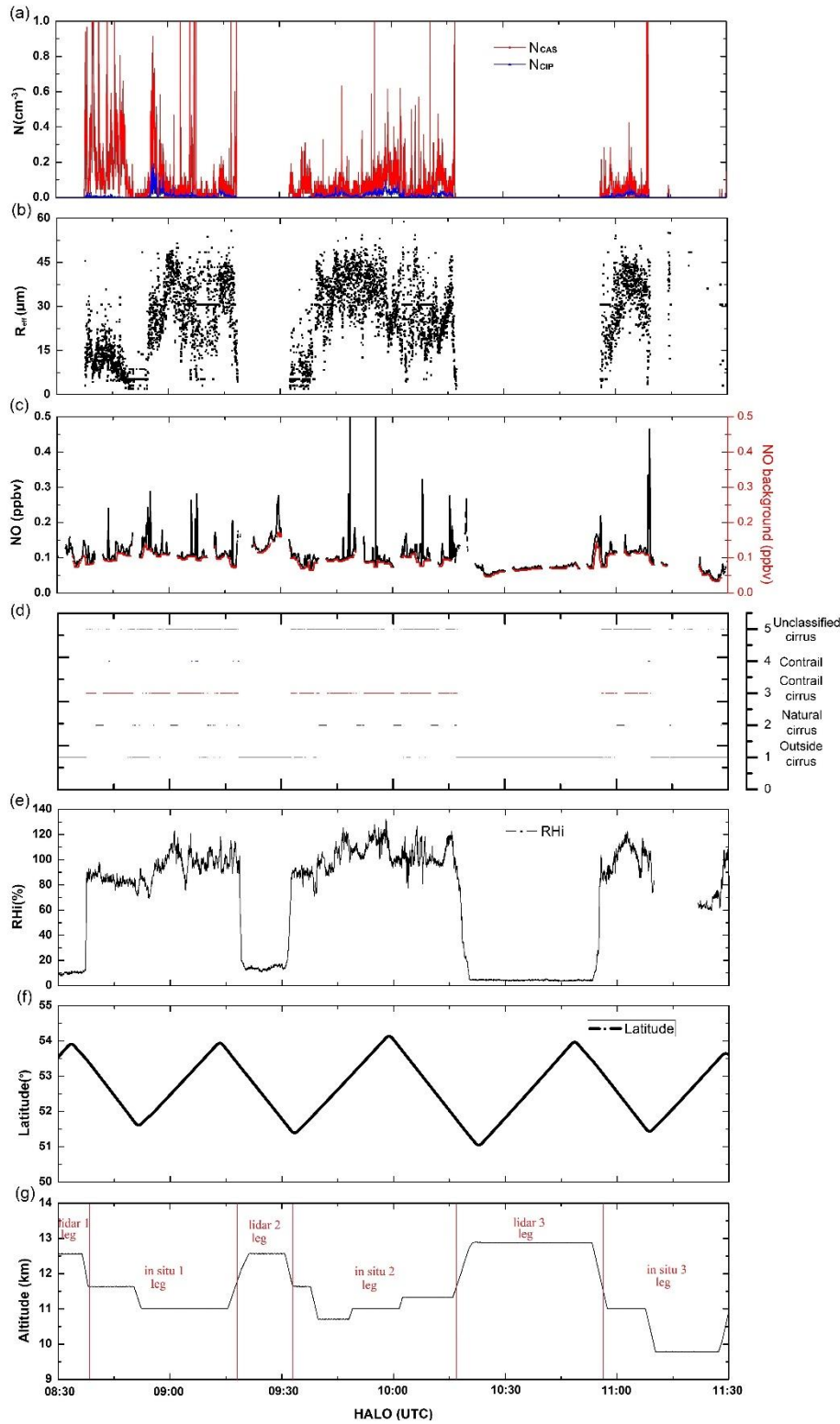



Figure 4: In situ measurements of HALO on 26 March 2014 over the North Atlantic region, including (a) ~~N~~ice number concentration N ~~> 3 µm~~, (b) R_{eff} ~~> 1.5 µm~~, (c) NO and NO background, (d) cirrus classification, (e) RHi, (f) flight latitude, and (g) altitude.

First, we determine dynamically the NO background concentration $NO_b(t)$ at a given time t by taking the minimum NO value in a 60-second sliding window around this time (Fig. 4c):

$$NO_b(t) = min_{-30s \leq \Delta t \leq 30s} NO(t + \Delta t) \qquad (1)$$

$NO_b(t)$ takes care of the natural variability of NO in this situation. Notably, we use the RANSAC algorithm (Fischler and Bolles, 1981) to interpret NO outliers and confirm that they haven't hit the NO background but the peaks of NO values. The difference between $NO_b(t)$ and the NO(t) at each time t is termed $\Delta NO(t)$

320
$$\Delta NO(t) = NO(t) - NO_b(t) \tag{2}$$

In a further step we consider ice number concentration N to classify contrails.

In accordance with Table 1, we first distinguish measurements in cirrus from "1. outside cirrus" when N_{CAS} and N_{CIP} are zero in step 1, and then separate "3. contrail cirrus" and "4. contrails" from "2. natural cirrus" when NO is higher than the background and N_{CAS} or N_{CIP} is larger than 0.03 cm^{-3} in step 2, reflecting the impact of air traffic. The rest is labelled as "5.
325   unclassified cirrus" in step 3. This class contains on one side those clouds that we cannot classify and likely consists mainly of natural cirrus. On the other side, ice cloud measurements with high ΔNO (> 0.14 ppbv) but moderate N ($N_{CAS} \leq 0.4$ cm^{-3}) do also fall in this category and should represent observations in younger plumes where few ice crystals could form (maybe because temperature is close to the Schmidt-Appleman criterion) or because ambient air is subsaturated leading to the evaporation of a considerable fraction of newly formed (small) ice crystals. Summarised, natural cirrus is identified when ice
330   crystals are present with either N_{CAS} or N_{CIP} larger than 0.03 cm^{-3} and NO close to the background value, i.e. $\Delta NO \leq 0.02$ ppbv; contrail cirrus is characterised by moderate values of NO, i.e. 0.02 ppbv < $\Delta NO \leq 0.14$ ppbv, and N_{CAS} or N_{CIP} > 0.03 cm^{-3}; contrails are assumed to consists of many small ice crystals, N_{CAS} > 0.4 cm^{-3}, and high NO peaks, i.e. ΔNO > 0.14 ppbv. The remaining cirrus is denoted as "unclassified cirrus".

Table 1: Cirrus classification according to microphysical properties (N), and tracer measurements (NO) measured by instruments
335   **aboard HALO.**

	Designation	N / cm^{-3}	ΔNO / ppbv	Note
1.	Outside cirrus	$N_{CAS} = 0$ and $N_{CIP} = 0$	Any value	Step 1
2.	Natural cirrus	$N_{CAS} > 0.03$ or $N_{CIP} > 0.03$	$\Delta NO \leq 0.02$	Step 2
3.	Contrail cirrus	$N_{CAS} > 0.03$ or $N_{CIP} > 0.03$	$0.02 < \Delta NO \leq 0.14$	Step 2
4.	Contrails	$N_{CAS} > 0.4$	$\Delta NO > 0.14$	
5.	Unclassified cirrus		The rest	Step 3

The determination of ΔNO and N thresholds are explained in S3 and Fig. S2. In total, from 08:30 to 11:30 UTC for each aircraft measurement with a frequency of 1 Hz we have classified 49 contrail observations, 1018 contrail cirrus observations and 2342 natural cirrus observations in Fig. 4d from in situ measurements. Unclassified cirrus encompasses 2472 cases, with
340   94 of them having ΔNO > 0.14 ppbv. At least 31% of all classified cirrus was in contrails or contrail cirrus, confirming the indications gained from MSG and WALES in Sect. 3.1 about the large number of contrails.

3.2.2 Effective radii

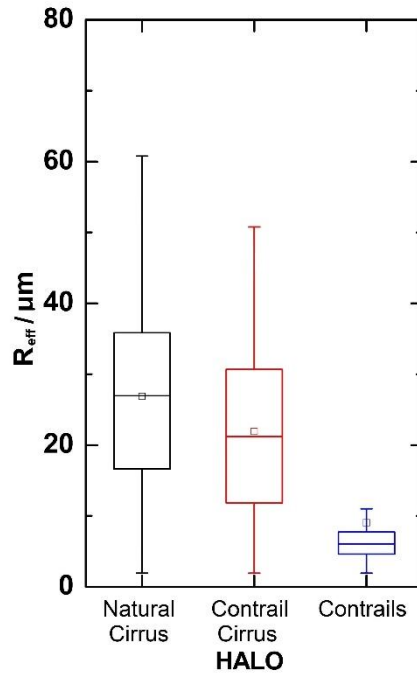



Figure 5: R$_{eff}$ of natural cirrus, contrail cirrus and contrails from HALO in situ measurements. Each bar contains the information of the maximum, minimum, interquartile range (25% percentile to 75% percentile), median (horizontal line) and mean (square) R$_{eff}$.

Considering the entire flight and separating R$_{eff}$ measurements according to our classification enables us to study the statistical properties of the ice crystal sizes. From Fig. 5, contrail cirrus R$_{eff}$ probed from airborne instruments are smaller than those of natural cirrus, with an average R$_{eff}$ of about 22 μm. The natural cirrus has an average R$_{eff}$ of 27 μm, which is at the lower end of the particle size distributions observed in natural cirrus (Schröder et al., 2000). The mean radii of contrails or contrail cirrus from in situ measurements is 18% smaller than that of natural cirrus. From contrails to contrail cirrus, the mean and medium R$_{eff}$ mainly show an increasing tendency by an average difference of 13 μm (144%) for this situation. Despite the uncertainty of probed particle sizes, the radii of natural cirrus reached up to 60 μm, while the majority of maximal R$_{eff}$ of contrail cirrus topped at 5~~13~~ μm, in agreement with findings of Voigt et al. (2017). This is consistent with the physical picture that contrails form initially as small particles and increase in size by water vapor uptake in ice supersaturated air. If no supersaturation is present, contrails dissipate. Still, contrail cirrus as well as natural cirrus can consist of sublimating particles when air becomes subsaturated (Kübbeler et al., 2011).

In sum, satellite, lidar and in situ measurements give combined evidence for an ideal case of a contrail cirrus outbreak with a high fraction of contrails and contrail cirrus embedded in natural cirrus. In the following, we investigate its radiative impact.

4 Radiative effects of contrail cirrus and natural cirrus

Here, we develop a new estimation method for TOA net instantaneous cirrus RF that relies on satellite and in situ observations. TOA irradiance of the contrail cirrus outbreak can be observed, but the comparable situation without ice clouds has to be obtained from another source, e.g. from the surroundings (like e.g. in Vázquez-Navarro et al., 2015 for contrails) or from model simulations (Haywood et al., 2009 for contrail cirrus). The instantaneous net RF at TOA is the change of the total irradiation under a situation with cirrus minus the irradiances in the same situation without cirrus. In fact, the SW component of the cirrus RF $RF_{SW_{TOA}}$ can be diagnosed as

$$RF_{SW_{TOA}} = SW_{\uparrow no\ cirrus} - SW_{\uparrow cirrus} .$$ (3)

Notice that $SW_{\uparrow cirrus}$ corresponds to the RSR that can be observed by the satellite. Similarly, the LW RF $RF_{LW_{TOA}}$ can be diagnosed from

370 $$RF_{LW_{TOA}} = LW_{\uparrow no\ cirrus} - LW_{\uparrow cirrus} . \tag{4}$$

Again, the cirrus term $LW_{\uparrow cirrus}$ in Eq. (4) corresponds to the OLR that can be computed from MSG. The net cirrus RF $RF_{NET_{TOA}}$ is defined as

$$RF_{NET_{TOA}} = RF_{SW_{TOA}} + RF_{LW_{TOA}} . \tag{5}$$

However, the two values (TOA irradiance with cirrus and without cirrus) are similar in size since contrail cirrus represent a
375 small perturbation. In fact, both in Vázquez-Navarro et. (2015) and Haywood et al. (2009) taking the difference between the cirrus contaminated and the cirrus-free irradiances leads also to negative values of RF in the thermal range (OLR with ice clouds is larger than OLR without ice clouds) or to positive RF in the solar range (RSR with ice clouds is smaller than RSR without ice clouds). This is unphysical and we would like to avoid it. To this end, in Sect. 4. 1 we developed a new method based on RTM calculations exploiting airborne measurements, satellite observations and ERA5 model atmospheric data that
380 produces TOA irradiance fully consistent for both cirrus and cirrus-free regions. So finally, to ensure that TOA irradiance calculated this way is realistic, we compare our RTM simulations of RSR and OLR for the cirrus contaminated case with satellite observations of RSR and OLR from RRUMS at pixel level. An additional comparison on a 3x3 pixel scale with the more accurate GERB products is performed to assess the RRUMS accuracy on this particular day (S4 and Fig. 3).

4.1 RTM simulations of TOA irradiance

385 In this section we illustrate the new method used to derive instantaneous cirrus RF and show that it compares well to observations of RSR and OLR by RRUMS. In particular, we explain a 2-step method to compute both cirrus-free and cirrus influenced TOA irradiances. First, an atmosphere is set up as input to the RTM that contains a realistic representation of the situation observed including in particular all clouds. Then in a second step the ice clouds are removed, for which the RF is to be computed. This provides the cirrus-free reference TOA irradiance for the calculation of cirrus RF. The basic setup of the
390 RTM is described in Sect. 2.3. Every RTM calculation needs an atmospheric state as input. This is in part obtained from ECMWF ERA5 reanalysis data (Hersbach et al., 2020). All 137 model levels are used. The horizontal and temporal resolution are $0.25° \times 0.25°$ and 1 h. We derived temperature profiles, logarithm of surface pressure, specific humidity, ozone mass mixing ratio, and land or sea mask. Densities of gaseous water (H_2O) and ozone (O_3) are derived from specific humidity and O_3 mass mixing ratio. CO_2 is set to a volume mixing ratio of 400 ppm. Vertical profiles of liquid clouds are also extracted from
395 ERA5 data. We choose vertical profiles close to the HALO flight time, i.e. with a time difference of 30 min at most between model and in situ measurements. We rely on the fact that reanalysis data should provide a realistic description of cloud properties and cloud positions, but we accept that this procedure might cause small temporal shifts such that observations close to cloud edges might not be represented in an optimal way through the model data. However, we refrain from interpolating cloud properties in time since also this procedure would create artificial clouds that do not exist in reality, especially in locations
400 where no cloud is present at a given time but it is there at the next time. For liquid clouds, the parameterization by Bugliaro et al. (2011, 2022) is applied for creating R_{eff} profiles using IWC and temperature from ERA5. The solar zenith angle corresponds to synchronous SEVIRI observations. Besides, the albedo of ocean is parameterized following Cox and Munk (1954a, b) and Nakajima and Tanaka (1983), especially involving the wind speed from ERA5. For ice clouds, another procedure is applied. Since SEVIRI observations with CiPS are able to account for the entire cirrus cloud layers but are only dependent on thermal channels and not affected by low lying clouds (Strandgren et al., 2017b), SEVIRI provides accurate ice cloud properties (IOT)
405 that can be used in the RTM.

The representation of cirrus at the flight level is complemented by adding their R_{eff} from in situ measurements, CTH and cirrus bottom height (CBH) from lidar legs, as well as CiPS IOT into libRadtran in the way described in the next lines. This way we simulated a vertically homogeneous ice cloud with the correct IOT obtained by CiPS. We assumed IOT to be constant over an

410 entire SEVIRI pixel while R_{eff} changes according to in situ information (at the altitude levels probed by HALO). Since the RTM needs IWC as input, we determine it from IOT. First, Extinction *Ext* for IWC = 1 g m⁻³ (i.e. extinction in km⁻¹/ (1 g m⁻³) for each measurement is interpolated to the given in situ R_{eff} with the parameterisation of optical properties of Baum et al. (201~~1~~4) for rough aggregates. The selection of this shape for all cloud types is motivated by the fact that each cloud column, even those containing contrails, encompass ice crystals with various temporal evolution, e.g. young small – probably round,

415 ice particles in contrail cores together with larger sedimenting ice crystals in the fall streaks with different shapes or with evaporating ice crystals – in part still large in size, in subsaturated air that are starting to lose asphericity or maybe even natural ice crystals with unknown shape. To avoid an additional arbitrary choice with respect to ice particle shape we decided to keep the method as simple as possible and selected this shape (rough aggregates) as e.g. for the MODIS optical property products Collection 6. A sensitivity study with respect to ice particle shape is conducted in Sect. 4.2. Note that the parametrization of

420 this shape only covers R_{eff} from 5 to 60 μm, which results in the inexecutable RTM calculations for larger or smaller ice crystals. 20 cases in total are removed but have a negligible effect on the estimation of radiative effects as R_{eff} of contrail cirrus and natural cirrus always fall in the range where RTM could simulate as indicated in Fig.5. The IWC for each measurement of R_{eff} corresponding to a vertically homogeneous ice cloud with given IOT is derived using the following equation:

$$IWC = \frac{IOT}{Ext*(CTH-CBH)} \tag{6}$$

425 CTH and CBH are obtained from the lidar legs since they seem to vary only slowly with time and space (Fig. 3). Thus, this IWC is used to simulate a homogeneous ice cloud layer between CBH and CTH and corresponds to the IOT observed by CiPS. Finally, in situ measurements of R_{eff} are assigned to a collocated IOT from CiPS as well as the ERA5 properties listed above for temperature, gas and liquid water clouds. With this atmospheric setup, TOA irradiances are computed and represent the cirrus contaminated RSR and OLR. Then, the ice clouds only are removed from the input of the RTM and other calculations

430 are performed to compute cirrus-free irradiances. Both together are then inserted in Eqs. (3)-(5) to compute the instantaneous net RF of the cirrus cloud under consideration.

The RSR and OLR values influenced by cirrus as obtained from libRadtran simulations (RSR_L and OLR_L) and compared with RRUMS algorithm outputs (RSR_R and OLR_R) along the flight path of HALO are presented in Fig. 6. The two methods agree quite well, but the RTM calculations tend to underestimate large RSR values, probably due to too thin, missing or even

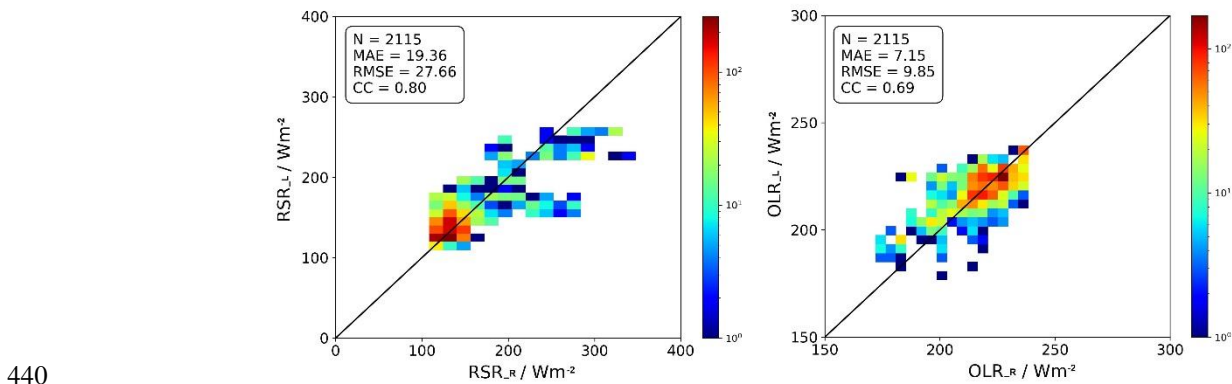
435 mismatched liquid water clouds that are taken from ERA5. Uncertainties in ice cloud properties cannot have such a large effect on RSR since their optical thickness is very low around 0.2. Furthermore, a smaller overestimation of RSR by the RTM compared to RRUMS is also observed for the smallest RSR values below 150 W m⁻², related to the bias of estimated ocean albedo but improved by the application of wind speed. For OLR, the agreement between the two datasets is good, with both slight (< 10 W m⁻²) overestimations and underestimations.

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Figure 6: Comparison of TOA (a) RSR and (b) OLR from our RTM simulations (RSR_L, OLR_L) for probed ice particles and RRUMS algorithm results (RSR_R, OLR_R) for single SEVIRI pixel along the HALO flight on 26 March 2014. The mean absolute error (MAE), root mean square error (RMSE) and correlation coefficient (CC) are used as metrics.

The RTM based TOA radiation estimation method provides a good representation of the real atmosphere and will be used to determine RF of the ice clouds in the following section.

4.2 Diurnal cycle of TOA RF of the contrail cirrus outbreak

In Sect. 4.1 we presented TOA RF for each waypoint of the probed cirrus. In order to examine microphysical properties and radiative effects of the contrail cirrus outbreak detected in this area and to analyse the corresponding temporal variation, we present the regional cirrus cover (CC), average cirrus IOT and CTH (all three quantities from CiPS), as well as mean TOA SW, LW and net RF in an area of 36×51 pixels ($\sim$16500 km^2, the red area in Fig. 2) near the HALO flight track, characteristic for contrail cirrus and cirrus as shown in Sect. 3. This area extends mostly east of the HALO flight and thus contains the contrails or contrail cirrus formed directly east of the ridge cloud. We consider here all ice clouds in the region with high aviation impact.

RF in this section is at the SEVIRI resolution, and calculated using CTH, IOT from CiPS, R_{eff} from in situ mean values of all cirrus (called $R_{eff,mean}$ in the following), and cloud thickness from WALES assuming the latter doesn't change in the area with the same procedure presented in Sect. 4.1 based on the RTM libRadtran.
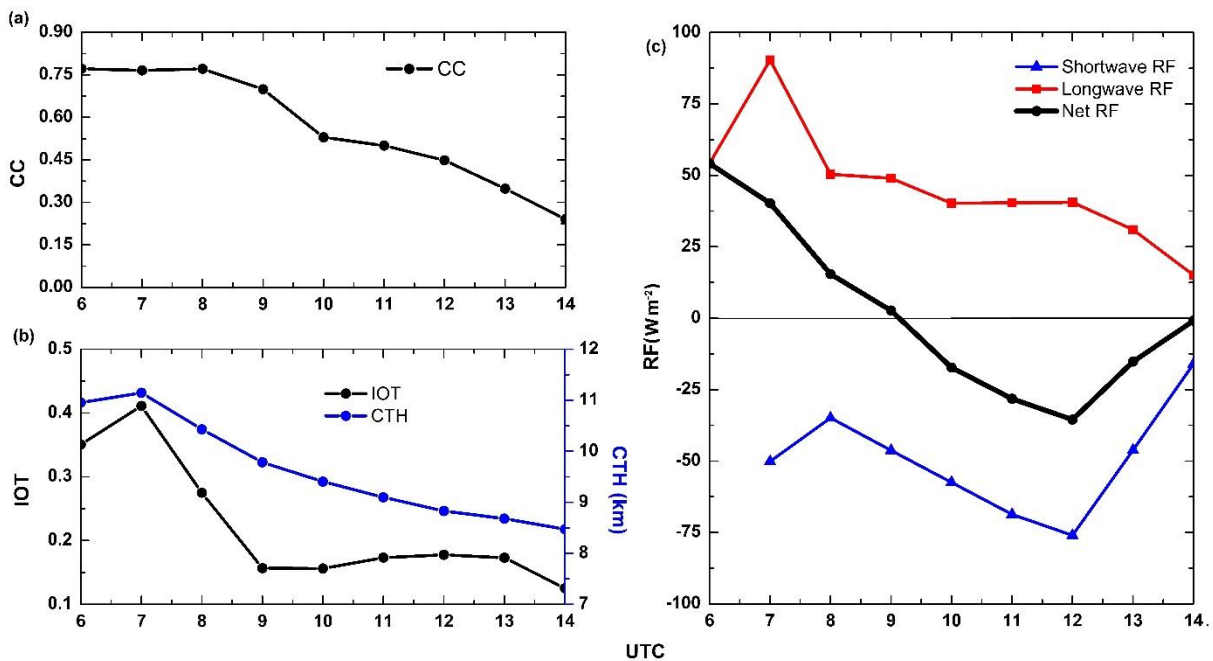
Figure 7: The variations of (a) CC, (b) mean IOT and mean CTH, and (d) SW, LW and net RF within the area indicated by a red box in Fig. 2.

In Fig. 7a, we observe that CC gradually decreases from 0.77 at 06 UTC to 0.25 at 14 UTC. The positive vertical velocity from ERA5 around that region implies the local downward motion of airmass to warmer temperature layers and the CTH also decreases. IOT in Fig. 7b decreases between 07:00 (0.41) and 10:00 UTC (0.15), then slowly increases until 12 UTC (0.17), then falls to 0.12. CTH decreases during the day and is thus consistent with both the observations of HALO (Fig. 3) and the downward motion. Since an underestimation of CTH by CiPS with respect to WALES (Fig. 3) is observed there, we assume that CTH is also underestimated by CiPS in this area.

Mean net RF over this area in this synoptic situation is positive in the early morning until 9 UTC with the maximum of net RF is at 7 UTC when the sun has risen. Hence the contrail cirrus outbreak is warming during night and early morning hours. After 9 UTC, the forcing becomes negative. More explicitly, from around 9 to 14 UTC the net RF is negative and thus this contrail cirrus outbreak tends to cool during daytime. The strongest cooling is observed at 12 UTC. Notice however that the possible underestimation of CTH by CiPS in this area would result in the general underestimation of the LW RF results since a lower CTH reduces the contrast to the cirrus-free OLR. In turn, this would further shift cirrus net RF towards cooling.

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}. Compared with aggregates, the ghm model has induced a larger net RF of 1.7Wm^{-2}, with the shift in SW and LW of 1.2 Wm^{-2} and 0.5 Wm^{-2}, respectively.

Table 2: 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.

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

5 Summary and conclusions

This study provides a detailed investigation of an ideal contrail cirrus outbreak event with airborne in situ probes, airborne lidar measurements and geostationary satellite observations using a case study focusing on the NAR on 26 March 2014 during the ML-CIRRUS experiment. We choose this contrail cirrus outbreak case because of the large contrail cirrus coverage and high air traffic density. As flight operation in all altitudes is not easily granted due to the high air traffic load in the NAR, the data presented here is also rare and unique in the sense that HALO was able to operate and acquire in-flight measurements of contrail cirrus perpendicular to the flight tracks of the NAR. From satellite remote sensing, few low-level water clouds and the relatively homogeneous oceanic background increase the sensitivity to retrieve cirrus properties. On the morning of that day the HALO research aircraft flew for three hours in or above cirrus clouds, contrail cirrus and contrails. Various contrails with related fall streaks have been identified in airborne lidar backscatter data which merge to a larger cloud with a vertical extent of 2 km that dissipates with time. High resolution RGBs and BTDs from MSG/SEVIRI also reveal the presence of various line shaped structures that represent contrails. Simultaneous airborne humidity measurements from AIMS show RHi between 80% and 120%, in line with lidar observations indicating a region where contrails formed, evolved and merged with natural cirrus. We identify aircraft plumes using peaks in NO data with an approach that takes care of the variable NO background, and contrast contrails and contrail cirrus on the basis of measured ice number concentrations. The mean radii of contrails or contrail cirrus is about 18% smaller than that of natural cirrus, suggesting a self-stabilizing mechanism of smaller contrail cirrus particle sizes surviving due to higher ice number concentrations in contrail cirrus compared to natural cirrus. Ice particle sizes increase by about 144% during the transition from contrail to contrail cirrus by deposition of water vapor in regions with ice supersaturation. Thus, combined evidence from satellite and aircraft data suggest the presence of an ideal contrail cirrus outbreak event.

For purpose of obtaining accurate radiative effects of contrail cirrus and natural cirrus, a new TOA RSR and OLR estimation method is developed, which is based on detailed RTM calculations and exploits in situ measurements, satellite observations and ERA5 model atmospheric data. Using IOT from MSG/SEVIRI (CiPS), R_{eff} from in situ, cloud top and bottom height from the lidar, and gas, temperature and liquid water cloud profiles from ERA5, an input atmospheric state for the RTM has been defined that enables to compute RSR and OLR that compare well to MSG measurements (RRUMS). When the ice cloud layer

505 is removed from the RTM input, RSR and OLR for cirrus-free conditions can be computed that are consistent with the corresponding cirrus data.

For a larger area of 36×51 SEVIRI pixels adjacent to the HALO flight path, the diurnal cycle of the contrail cirrus outbreak was computed. RF of cirrus is associated with the changes of cirrus coverage, CTH and IOT. Here, we find a positive net RF of the contrail cirrus outbreak region in the early morning, and the contrail cirrus outbreak warms at TOA till 9 UTC. Then,

510 during daytime the mean net RF of the contrail cirrus outbreak becomes negative, and the contrail cirrus outbreak cools. In the long-term observations or hourly resolved simulations of contrail cirrus coverage and RF in the NAR, Graf and Schumann (2012), Duda et al. (2013), and Vázquez-Navarro et al. (2015) also find the contrail coverage is important and that a high variability of the contrail impact exhibits. These studies indicate that contrail cirrus warm during the night, while a larger variability in contrail cirrus RF exists during the day. Teoh et al. (2022a, b) show that contrail cirrus often cools during daytime

515 and that the cooling depends on many parameters including solar zenith angle and the surface albedo. For some cases with low clouds Teoh et al. (2022a, b) also find warming contrail cirrus during the day. In the absence of sunlight during night time however, contrail cirrus warms the atmosphere. Here we use a new method to derive the RF of contrail cirrus and cirrus and we test it using in-situ and lidar observations in a contrail cirrus outbreak situation. A following study will aim at applying the method to a broader data set to investigate the radiative effects of cirrus and contrail cirrus using RTM simulations involving

520 CTH and IOT from satellite, R_{eff} from in situ values, and CBH and cloud thickness from lidar.

This work is valuable for identifying contrails, contrail cirrus and natural cirrus from different platforms, estimating TOA RF from satellite data, assessing microphysical properties and climate impacts of anthropogenic cirrus and natural cirrus, and can help to formulate appropriate contrail mitigation options.

525 *Data availability*

Flight measurements are available at the HALO data base at https://halo-db.pa.op.dlr.de/. The SEVIRI data are provided by EUMETSAT (European Organisation for the Exploitation of Meteorological Satellites) and the modelled atmospheric profiles are obtained from ECMWF (European Centre for Medium-Range Weather Forecasts). The GERB data could be accessed from EUMETSAT (GERB international team).

530

Author contributions

Z.W. conducted the analysis and wrote the manuscript. L.B. advised the study and provided feedbacks on the manuscript. L.B., M.W., and G.D. plotted Fig. 1 & 2, 3, and S1. C.V. coordinated the ML-CIRRUS mission and L.B. and T. J. contributed to the flight planning. T.J., R.H., H.Z., M.W., S.G., S.Ka., and C.V. participated in the flight measurements. G.D. wrote the

535 description of WALES datasets. Z.W., L.B., T.J., R.H., H.Z., and C.V. discussed the cirrus classification. U.B. helped with the interpretation of the cirrus cloud conditions and evolution. All authors contributed to and commented on the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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