CIRRUS-HL: Picturing high- and mid-latitude summer cirrus and contrail

cirrus above Europe with airborne measurements aboard the research

aircraft HALO

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ABSTRACT: Accurately determining and reducing the climate impact of aviation and its uncertainties is one of the pressing challenges of our times. Contrail cirrus are estimated to contribute more than half of the total effective radiative forcing from aviation, yet the uncertainties in their optical and radiative properties are large. In contrast to mid-latitude cirrus, high-latitude cirrus are less anthropogenically influenced, thus they are more pristine. However, little is known about Arctic cirrus properties and their role in the amplified warming of this region. The CIRRUS-HL (Cirrus in High Latitudes) mission using the High Altitude and LOng range research aircraft HALO provides measurements in mid and high latitudes during the summer (June/July) 2021, exploiting HALO's capabilities and a comprehensive cloud-aerosol-trace gas and radiation instrumentation. The results of 24 HALO flights provide new insights into both natural cirrus and contrail cirrus properties in high (60°N-76°N) and mid latitudes (38°N-60°N). In particular, we find lower ice water content (-42%) and lower number concentrations (-88%) of cirrus particles with larger mean diameters (+22%) in high latitudes. Ice supersaturated regions were frequently observed in mid and high latitudes, with median in-cloud relative humidity over ice between 105% and 122%. Mean aerosol number concentrations in the mid latitudes were reduced by up to 80% compared to pre-COVID times. Less air traffic during the COVID-19 lock-downs, reduced contrail cirrus coverage and lower ice nucleating particle concentrations in high latitudes help to explain the observed differences in cirrus properties. The extensive data set will be used to improve weather and climate models.

SIGNIFICANCE STATEMENT: In contrast to Arctic cirrus, mid-latitude cirrus are more often modified by human activities, of which air traffic is a significant contributor through formation of contrails and contrail cirrus. These man-made cirrus warm the Earth, but to constrain their effects on climate, in-situ and remote sensing measurements were conducted with the German research aircraft HALO. During 24 flights, we used HALO's exceptional altitude and distance range to sample and contrast different cirrus types from the dense air traffic regions to the remote Arctic regions. We find that microphysical properties of high- and mid-latitude cirrus differ substantially, related to their formation pathway, the abundance of air traffic and the availability of ice nucleating particles. The measurements will help to validate contrail cirrus and climate models.

CAPSULE: Cirrus clouds and contrail cirrus were probed in summer 2021 during the airborne HALO mission CIRRUS-HL, contrasting aerosol and humidity distributions, microphysical and optical ice crystal properties in high- and mid-latitudes.

1. Introduction and scientific background

Cirrus clouds are ice clouds that exist in the upper troposphere at temperatures below -38°C. Air masses that are lifted cool and eventually reach supersaturation, which leads to ice formation below this temperature. Cirrus clouds exert an overall net warming effect on the atmosphere-surface system, where the warming effect through absorption of thermal radiation exceeds the cooling effect through reflection of sunlight (Chen et al. 2000). The representation of cirrus clouds in general circulation models comprises a main challenge and a major uncertainty for predicting the rate and geographical pattern of climate change (Gasparini et al. 2018; Zelinka et al. 2020). Midlatitude cirrus clouds are formed in various synoptic situations with different dynamical forcings. The interplay of updraft and associated cooling with the availability of ice nucleating particles (INPs) determines whether the cirrus cloud forms homogeneously or heterogeneously (Kärcher and Lohmann 2002, 2003) and whether it forms in situ or via the liquid phase (Wernli et al. 2016; Luebke et al. 2016; Krämer et al. 2016; Voigt et al. 2017; Krämer et al. 2020). Cirrus that form at temperatures below 235 K, heterogeneously through deposition of water vapor onto preexisting INPs or homogeneously through ice nucleation in supercooled solution particles, are referred to as in situ origin cirrus. Liquid-origin cirrus form through the freezing of existing liquid cloud droplets, either heterogeneously at temperatures above 235 K via embedded INPs

or homogeneously at 235 K. Cirrus clouds can exert a positive or a negative radiative forcing (RF) depending on a number of factors like microphysical (e.g. ice crystal size, number) and macrophysical (e.g. location, time) properties. Polar night cirrus tend to warm the atmosphere due to exclusive absorption of terrestrial radiation (Hong and Liu 2015; Marsing et al. 2023). The radiative effects of the polar-day Arctic and mid-latitude cirrus depend on their optical thickness, ice crystal optical and microphysical properties, the surface albedo and emission, the vertical extension and location of the cloud, and the solar zenith angle (Krämer et al. 2020; Hong and Liu 2015; Lelli et al. 2023), thus their effect is complicated to determine. Similar to Arctic low-level clouds and their relation to the accelerated warming of the Arctic (McFarquhar et al. 2011; Wendisch et al. 2017, 2023), the net contribution of cirrus clouds to the Arctic temperature trend is unknown. A better understanding of their influence on radiative balance (Marsing et al. 2023) and temperature in this sensitive region of the Earth requires observational data of ice microphysical and optical properties, water vapor and aerosols from the ground to the lower stratosphere, which is hard to obtain due to the limited accessibility of this region (Kay et al. 2016; Schmale et al. 2021).

Recent studies have shown that anthropogenic cirrus clouds induced by aviation (mainly contrails, and the contrail cirrus that evolve from them) contribute substantially to mid-latitude upper tropospheric clouds with an expected warming effect (Minnis et al. 2004; Burkhardt and Kärcher 2011). Current aviation climate impact assessments conclude that contrail cirrus have a net effective radiative forcing with a magnitude that exceeds the effect of the integrated CO₂ emissions from aviation since the beginning of modern aviation (Lee et al. 2021). Mitigation approaches to reduce the impact of contrails include using sustainable aviation fuels (Voigt et al. 2021; Märkl et al. 2024), technological means (Gierens 2021; Bier et al. 2024), rerouting of aircraft to avoid climate sensitive regions (Teoh et al. 2022a; Ng et al. 2024) or a combination of these measures (Teoh et al. 2022b).

A large uncertainty for the climate impact assessment of contrail cirrus is the prediction of ice supersaturated regions (ISSRs) over the lifetime of the contrail – a fundamental requirement to assess its radiative forcing (Jensen et al. 2001; Gierens et al. 2020). High-resolution measurements of low water vapor concentrations in the upper troposphere and lower stratosphere (UTLS) have improved significantly in the past years (Thornberry et al. 2013; Rollins et al. 2014). These measurements revealed a significant model offset in current weather prediction data in the UTLS

(Kaufmann et al. 2018; Krüger et al. 2022). Furthermore, uncertainties in the representation of ice crystal shape (Wendisch et al. 2005, 2007) and complexity of high- and mid-latitude cirrus and contrail cirrus (Järvinen et al. 2018; Chauvigné et al. 2018), in the modification of optical properties of natural clouds through contrail cirrus (Tesche et al. 2016; Marjani et al. 2022), and in their life cycle and variability (Unterstrasser et al. 2017; Bier and Burkhardt 2022) are limiting factors to the global climate impact assessment of these man-made clouds. High-resolution upper tropospheric measurements of water vapor and clouds are therefore key to understand the local and global effects of natural and anthropogenic cirrus (Sassen 1997; Heymsfield et al. 2017; Groß et al. 2022), and finally their climate feedback (Bock and Lauer 2024).

Here, we present an overview of the measurements performed using the High Altitude and LOng range research aircraft HALO in the frame of the Cirrus in High Latitudes (CIRRUS-HL) mission (www.cirrus-hl.de), which focused on mid- and high-latitude cirrus clouds, water vapor, and aerosols, as well as contrails and contrail cirrus during summer 2021. We provide an overview of the scope of the CIRRUS-HL campaign, the instruments, and the forecast products used for flight planning. We elaborate on the 24 flights performed between 38°N and 76°N and examine the latitudinal differences observed in the cloud, water vapor, trace gas, and aerosol data. Furthermore, selected cases are used to demonstrate the capabilities and application of the data set by combining measurements from state of the art airborne instrumentation with model data and satellite retrievals.

2. Scope of the CIRRUS-HL mission and atmospheric conditions during campaign period

The aim of the CIRRUS-HL campaign was to probe optical, radiative, micro- and macrophysical properties of cirrus clouds from the mid-latitude, dense air traffic regions to the high-latitude more pristine Arctic regions to investigate the following topics:

- How do the microphysical and radiative properties of cirrus clouds in Arctic regions differ from those of mid-latitude cirrus clouds? Is there a distinguishable anthropogenic effect at mid latitudes? What is the relative contribution of in situ and liquid-origin cirrus?
- Are there differences in aerosol and water vapor distributions at high and mid latitudes?
- Do contrails and contrail cirrus significantly modify microphysical, optical and radiation properties of cirrus at mid latitudes?

• Are cirrus clouds and contrail cirrus adequately represented in climate models? What are the dominant ice nucleating particles of high- and mid-latitude cirrus clouds? What is the role of cirrus clouds for the Arctic amplification?

The mission flights were conducted from Oberpfaffenhofen (Southern Germany), with refueling stops in Scandinavia or Iceland for the high-latitude cirrus missions. We refer to measurements as high latitude when the aircraft was North of 60°N latitude. Sensitivity studies through variation of this latitude threshold by 5° did not substantially impact the result of the analysis (De La Torre Castro et al. 2023).

The CIRRUS-HL mission was conducted during a time when air traffic was still reduced due to the COVID-19 pandemic. According to EUROCONTROL, the number of daily flights was increasing sharply during the months of June and July 2021, but on average still 65% to 80% lower than in the same months of 2019. This led to reduced global contrail and contrail cirrus coverage, resulting in a 50% reduced RF from contrail cirrus compared to pre-COVID 2019 (Voigt et al. 2022; Schumann et al. 2021b,a). Thus, the CIRRUS-HL measurements were performed in a time with reduced effect of aviation on the cirrus coverage and radiative forcing.

The high-latitude cirrus flights were focused on thin cirrus clouds, with air mass origins (based on 10-days backward trajectories) in the Northern latitudes (> 60°N). Sea ice was at an interannual minimum during the campaign period, with less than 50% of the maximum extension in March 2021, providing cirrus measurements mainly above open ocean and above land with a high occurrence frequency of low-level clouds. In addition, July 2021 was affected by a quasi-stationary low pressure system residing above central Europe. The enhanced heat and moisture transported from the Baltic sea caused strong convective activity and catastrophic floods in Europe, particularly in Germany and Belgium (Lehmkuhl et al. 2022), with an unknown effect on the humidity fields at cirrus altitude (Krüger et al. 2022).

3. Overview of flights

In total, 24 flights (including 23 scientific flights and 1 test flight) were conducted between 38°N and 76°N. Sampling 34 flight hours (17.7 h in mid-latitude cirrus, 7.8 h in high-latitude cirrus, and the remaining time in mixed-phase and liquid clouds), provided in situ measurements at temperatures down to -63°C and altitudes up to 14.3 km. A list of the research objectives of

the individual flights, and the altitude, geographical and temperature ranges sampled, is presented in Table 1. An overview of the horizontal and vertical flight paths of the individual flights is presented in Fig. 1 and the supplement movie. We also provide an overview on the number of measurements in liquid-origin and in-situ cirrus per 2°-latitude band. The classification of these cirrus types is based on liquid water content (LWC) and ice water content (IWC) predicted by the European Center for Medium-Range Weather Forecast (ECMWF) data along 10-days backward trajectories calculated with LAGRANTO (Sprenger and Wernli 2015). Starting from the HALO flight paths, the hourly backward evolution of the IWC and LWC along the trajectories was evaluated to estimate the age of the cloud and distinguish between in situ and liquid origin cirrus. If liquid water was present along the trajectory between cloud formation and the measurement point, we refer to the cloud as a liquid-origin cirrus following the approach described in Wernli et al. (2016) and Luebke et al. (2016) and for CIRRUS-HL data explained in more detail in De La Torre Castro et al. (2023).

Next to thin jet stream cirrus, liquid-origin cirrus in high and mid latitudes were sampled, contrail cirrus cluster, day-and-night-time radiative effects of cirrus, cirrus formed on aviation soot were investigated, and dedicated measurements of cloud vertical profiles in convective cirrus were performed (see Supplement). With the exception of two flights focusing on convective cirrus, mostly synoptically-forced cirrus were targeted. Cirrus as well as mixed-phase clouds were also targeted above ground-based observational sites like the radar station of the German Weather Service in Hohenpeißenberg (HP) and the lidars and ceilometer at the TROPOS institute in Leipzig and the Meteorological Institute Munich (MIM).

Contrail cirrus were detected in-situ as isolated clusters, above cirrus and embedded in cirrus, with co-located remote sensing measurements (see Section 7c). In total more than 600 contrail cirrus encounters with contrail ages between 100 and 10000 s were detected in situ during the campaign. Based on this analysis, a contrail cirrus-flag was derived that identifies sequences of individual contrail cirrus encounters using coincident measurements of NO and aerosol number in the data set. A class of contrail cirrus with high ice crystal number ($N_{ice} > 0.1 \text{ cm}^{-3}$) and low effective diameter (ED < 40 µm) was identified in the in-situ data (De La Torre Castro et al. 2023). We discuss the effect of contrail cirrus on upper tropospheric humidity in Section 6c and on radiation in Section 7d. An overview of the cloud microphysical properties is given in Section 6.

So-called missed approach flight patterns were repeatedly performed above European cities during the campaign to randomly capture the aerosol, cloud and trace gas vertical profiles in dense air traffic and remote regions. These vertical profiles provide information on the aerosol background during the campaign for comparisons with models, i.e. EMAC (Kaiser et al. 2019; Beer et al. 2020; Righi et al. 2020) and MECO(n) (Mertens et al. 2016) and relate atmospheric conditions to previous aerosol and trace gas conditions in pre-COVID and lock-down conditions e.g. during BLUESKY (Voigt et al. 2022; Tomsche et al. 2022) (see Section 6b).

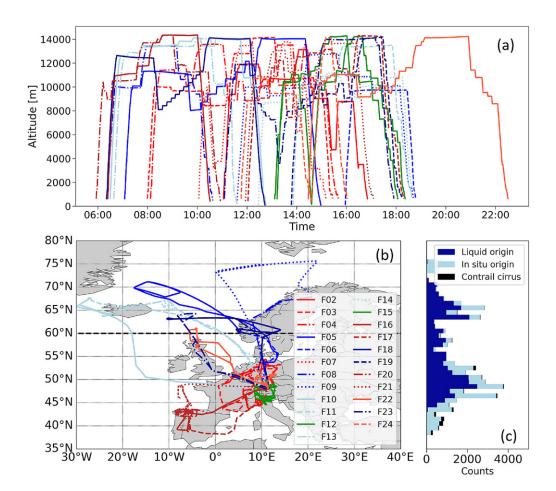


Fig. 1. Overview of region, time and altitude of CIRRUS-HL scientific flights. (a) GPS Altitude profile of 23 CIRRUS-HL flights with respect to the time of the day (UTC). High altitude flight legs were dedicated to remote sensing measurements. Cirrus were probed in-situ with staircase flight pattern ascending through the clouds. (b) Map with flight paths of 23 flights. Flights at high latitudes are indicated with blue colours, red colours represent flights at mid latitudes, and green colors indicate flights in convection. The 60 degree of latitude is indicated with a thick dashed line. (c) Number of cloud encounters classified as either liquid origin, in situ origin or contrail cirrus per 2°-latitude interval. Contrail cirrus were mainly found above continental Europe.

TABLE 1. Summary of the CIRRUS-HL science flights in June and July 2021 including objectives, flight times, in-cloud and clears-sky sampling times (below 235 K), latitude, and longitude ranges. Measurement altitude and temperature ranges are indicated only for the cirrus regime measurements. For flights with targets above ground-based stations like the Leibniz Institute for Troposphärenforschung (TROPOS) in Leipzig, Meteorological Institute Munich (MIM) and Hohenpeißenberg (HP), ground-based radar or lidar data are available.

Flight	Date	Flight duration	In-cloud / Clear-sky	Latitude [°N]	Longitude [°W]	Altitude [km]	Temperature [°C]	Targets
F02	25 June 2021	5 h 48 min	42 min / 3 h 55 min	[43.6,54.3]	[6.0, 13.4]	[8.5, 9.8]	[-49.5, -38]	ML cirrus above Germany, embedded contrails (Section 7c)
F03	26 June 2021	7 h 28 min	1 h 40 min / 4 h 12 min	[43.1, 52.4]	[0.5, 12.5]	[8.8, 11.4]	[-56,-38]	ML cirrus, embedded contrails, overpass Leipzig, MIM/HP
F04	28 June 2021	7 h 34 min	2 h 5 min / 3 h 46 min	[43.7,53.7]	[6.0, 13.3]	[9.2, 12.2]	[-60.9, -38]	ML frontal cirrus, embedded contrails, overpass Leipzig, MIM/HP
F05 / F06	29 June 2021	10 h 43 min	3 h 15 min / 5 h 25 min	[47.8,71.1]	[-17.5, 13.6]	[9.2, 12.5]	[-62.7, -38]	HL (Iceland and Norway) and ML cirrus, embedded contrails; overpass MIM/HP
F07	01 July 2021	5 h 7 min	1 h 1 min / 2 h 5 min	[47.6,53.4]	[7.8, 20.7]	[8.7, 11.3]	[-55.9, -38]	ML cirrus, embedded contrails
F08 / F09	05 July 2021	11 h 22 min	2 h 21 min / 7 h 21 min	[48.0, 75.6]	[0.1, 22.0]	[9.2, 11.6]	[-57, -39.6]	HL cirrus (Norway)
F10/F11	07 July 2021	11 h 5 min	1 h 17 min / 8 h 13 min	[47.9,67.7]	[-29.8, 11.4]	[10, 13.8]	[-54.3, -44.2]	HL cirrus (Iceland), contrails, BB, (Section 8)
F12	08 July 2021	4 h 51 min	1 h 33 min / 1 h 53 min	[45.3, 49.1]	[8.4, 13.2]	[9,11.7]	[-53.2, -38]	ML cirrus, convection over the Alps, (Supplement)
F13/F14	12 July 2021	11 h 16 min	3 h 18 min / 5 h 47 min	[47.7,67.0]	[-24.4, 11.6]	[8.8,11.7]	[-55.1, -38]	HL and ML cirrus, contrails, (Section 5)
F15	13 July 2021	5 h 15 min	1 h 30 / 1 h 44 min	[44.6,49.9]	[9.5, 12.9]	[9.5, 12]	[-53.8, -38]	ML cirrus, convection, dust
F16/F17	15 July 2021	10 h 23 min	1 h 31 min / 6 h 16 min	[38.2, 48.2]	[-8.7, 11.7]	[8.7, 14.3]	[-61.6, -38]	in situ origin ML cirrus, contrail outbreak
F18/F19	19 July 2021	10 h 49 min	1 h 31 min / 6 h 3 min	[48.1, 63.4]	[-10.4, 11.4]	[9.1, 11.8]	[-60, -38]	HL cirrus, contrails, soot cirrus
F20 / F21	21 July 2021	10 h 9 min	1 h / 7 h 28 min	[42.0,49.0]	[-8.4, 11.4]	[9.9, 13.6]	[-56.4, -41.1]	in situ origin ML cirrus, contrails, CALIPSO overpass, (Section 7c)
F22	23 July 2021	8 h 2 min	1 h 3 min / 5 h 51 min	[48.1,61.1]	[-5.7, 11.4]	[9.2, 11.5]	[-57.2, -38]	HL cirrus, day-night, embedded contrails
F23	28 July 2021	6 h 49 min	41 min / 3 h 49 min	[48.0,64.3]	[-8.2, 11.4]	[7.5,11.9]	[-52.9, -38]	HL cirrus and ML, CALIPSO overpass
F24	29 July 2021	4 h 16 min	- / 2 h 27 min	[48.0,51.8]	[8.3, 13.1]	1	1	nose boom turbulence calibration

4. HALO-Instrumentation

The HALO instrumentation for CIRRUS-HL is based on the payload of the previous in-situ cloud mission ML-CIRRUS (Table 1 and 2 in Voigt et al. (2017)). It comprised a comprehensive and extended payload to characterize ice particle residuals (IPR), aerosol size, number and composition, INP concentration, cirrus ice crystal microphysical and optical properties, radiative fluxes and humidity in the tropopause region. A list of cabin-based instruments and underwing cloud-probes with the measured quantity, respective size range and acronym is given in Table 2 and Table 3. Fig. 2 provides pictures of instruments central to this study and of new instruments deployed for the first time on HALO. One significant improvement to the previous mission is the new mounting position of the counterflow virtual impactor (Ogren et al. 1985) for HALO (HALO-CVI) at the lower fuselage (Fig. 2a), as cloud particle enrichment and shadow effects are significantly lower compared to sampling at the upper fuselage. This cloud inlet sampled cirrus ice particles and provided their IPR to a number of aerosol instruments. This mainly includes two ultra-high sensitivity aerosol spectrometers (CVI-UHSAS and FASD-UHSAS (Cai et al. 2008)), the singleparticle laser ablation mass spectrometer ALABAMA (Brands et al. 2011; Clemen et al. 2020), the SOPAMA system including a single particle soot photometer extended range (SP2xr, Droplet Measurement Technologies) instrument, two tricolor absorption photometers (TAP, Brechtel Inc), as well as the sampler HERA4HALO (Grawe et al. 2023). Most of the aerosol instruments connected to the HALO-CVI were alternating also operated behind the HALO Submicrometer Aerosol Inlet (HASI). Interstitial aerosol measurements were affected by cloud hydrometeors and therefore aerosol measurements with the HASI inlet were only used in cloud free conditions. Fig. 2b shows the HERA4HALO sampler, deployed for the first time on HALO during CIRRUS-HL. The sampler allows for up to six filters being selectively sampled during one research flight. After flight, filters were removed from their holders, kept frozen in a freezer at -20°C until used for offline immersion INP measurements (see e.g. (Grawe et al. 2023)). Fig. 2c shows the Backscatter Cloud Probe with Depolarisation (BCPD) installed on the second window on the right side of the aircraft. The BCPD detects particles in the size range between 2 and 42 µm through light scattering in the 155° backward direction (Lucke et al. 2023). The BCPD was deployed on HALO for the first time for comparison to underwing-cloud-probes. Backscatter lidar and water vapor DIfferential Absorption Lidar (DIAL) profiles were recorded using the airborne demonstrator for the WAter

vapour Lidar Experiment in Space (WALES) (Wirth et al. 2009; Groß et al. 2014) (see Fig 2d). The data set gives time series of vertical profiles of backscatter ratio, particle depolarization and water vapour molecular density measured along the flight path of HALO. Backscatter ratio and aerosol depolarisation data are given with one second time resolution and 15 m vertical resolution at a wavelength of 532 nm. The backscatter profiles are extinction corrected using the High Spectral Resolution Lidar (HSRL) method (Esselborn et al. 2008). The water vapour profiles have a time resolution of 12 s and a vertical binning of 15 m. As the DIAL measures the molecular density of water vapour, temperature/pressure data have to be used to convert this into mixing ratio or relative humidity. For this purpose ECMWF analyses are interpolated in space and time to the lidar curtain.

A new element of the CIRRUS-HL payload was the spectrometer of the Munich Aerosol Cloud Scanner (specMACS; Ewald et al. 2016; Weber et al. 2024) shown in Fig. 2e. It is a high spatial resolution hyperspectral and polarized imaging system and operated in a downward-looking perspective. The instrument consists of two hyperspectral cameras with a field of view of 32.7° and 35.5°, respectively, which are sensitive to the wavelength range between 400 and 2500 nm. In addition, the spectrometers are complemented by two 2D RGB polarization-resolving cameras with a maximum combined field of view of 91°×117°. The radiative energy budget at flight altitude was measured with the Broadband AirCrAft RaDiometer Instrumentation (BACARDI, Ehrlich et al. 2023) and the Spectral Modular Airborne Radiation measurement sysTem (SMART, Wolf et al. 2019). BACARDI consist of two sets of pyranometers and pyrgeometers to cover the upward and downward solar (0.3–3 µm) and thermal-infrared (3–100 µm) irradiances. This combination allows net irradiances to be quantified and with help of radiative transfer simulations to calculate the cloud radiative effect as described by Luebke et al. (2022). Spectrally resolved solar irradiances (upward and downward) are measured by SMART in the wavelength range between 0.3 to 2.1 µm (2–10 nm spectral resolution). In addition, a suite of trace gas measurements including ozone (O₃) from the Fast AIRborne Ozone Instrument (FAIRO) (Zahn et al. 2012), nitrogen oxide and the sum of reactive nitrogen (NO and NO_v) from the AENEAS (AtmosphEric Nitrogen oxides mEAsuring System) instrument (Ziereis et al. 2022) together with carbon dioxide (CO₂) and methane (CH₄) measured with a Picarro instrument (Klausner 2020) provided important information on the tropospheric and stratospheric air mass composition. Water vapor was measured using several in-situ instruments: The FISH (Schiller et al. 2008; Meyer et al. 2015), the AIMS (Kaufmann 2016; Jurkat et al. 2016),

the SHARC (Kaufmann et al. 2018) and the WARAN instrument (Marsing et al. 2023). The measurement capabilities of these instruments are documented in Kaufmann et al. (2018).



Fig. 2. Selected instruments (newly) deployed on HALO during CIRRUS-HL: (a) CVI mounted at the lower fuselage next to BARCARDI. In the background, the wingpods for the PIP, SID-3 and CAS-DPOL are visible. (b) Picture of the HERA4HALO sampler. (c) Picture of the BCPD and mounting position on HALO at the second window on the right. (d) Picture of the cabin with the HSRL lidar WALES. (e) Picture of the specMACS camera system. (f) HALO rack with aerosol and the trace gas instrumentation (SP2xr, TAP, UHSAS and PICARRO) (g) Picture of the right wing carrying (from outside position to inside) the PHIPS, the CCP, the NIXE-CAPS and the MTP during cirrus and contrail sampling at sunset. Acronyms are explained in Table 2 and 3.

Table 2. Underwing cloud probes deployed on HALO during CIRRUS-HL

Instruments	Measured Properties, Range	PI, Institution	Reference
CCP Cloud Combination Probe	Cloud particle size distribution, 2 - 960 µm	Voigt, Uni Mainz	De La Torre Castro et al. (2023)
PIP Precipitation Imaging Probe	Cloud particle size distribution, 100 - 6400 µm	Voigt, Uni Mainz	De La Torre Castro et al. (2023)
NIXE-CAPS Cloud and Aerosol Spectrometer	Cloud particle size distribution, 0.5 - 960 µm	Krämer, FZJ	Krämer et al. (2020)
SID-3 Small ice detector Mark 3	Ice particle size and phase, $2-50 \mu m$	Schnaiter, KIT	Vochezer et al., (2016)
PHIPS Particle Habit Imaging and Polar Scattering Probe	Ice particle habit and phase function, 10-1000 µm	Järvinen, KIT	Schnaiter (2016)
CAS-DPOL Cloud and Aerosol Spectrometer	Ice particle size distribution, 0.5 - 50 μm	Jurkat-Witschas, Voigt, DLR	Voigt et al. (2021)
PCASP-100X Passive Cavity Aerosol Spectrometer Probe	Dry particle size distribution, 0.12-3,5 µm	Sauer, DLR	Voigt et al. (2022)
MTP Microwave Temperature Profiler	temperature profiles	Tomsche, DLR	Heckl et al. (2021)

TABLE 3. Cabin-based instruments deployed on HALO during CIRRUS-HL

Instruments	Measured Properties, Range	PI, Institution	Reference
WALES high spectral resolution lidar with H2O differential absorption channel	water vapor concentrations, Backscatter ratio and polarization	Wirth, DLR	Wirth et al. (2009)
Albedometer for Spectral Modular Airborne Radiation measurements System (SMART)	Spectral radiance, spectral irradiance 350-2200 nm	Wendisch, Uni Leipzig	Ehrlich et al. (2008)
Broadband AirCrAft RaDiometer Instrumentation (BACARDI)	upward and downward solar (0.3–3 µm) and thermal–infrared (3–100 µm) irradiances	Giez, Wendisch, DLR, Uni Leipzig	Ehrlich et al. (2023)
Counterflow Virtual Impactor sytem (HALO-CVI) inlet with CPC/UHSAS/PSAP/WVSS-II	Cloud particle residual number concentration, size distribution (0.65 nm to 1 µm), absorption coefficient, IWC	Mertes, TROPOS	Cai et al. (2008); Mertes et al. (2004); Vance et al. (2015)
High-volume flow aERosol particle filter sAmpler (HERA)	aerosol particle sampling for offline immersion INP analysis between 0° C to -30° C freezing temperature	Stratmann, TROPOS	Grawe et al. (2023)
Aerosol Measurement System with condensation particle counter (CPC)/optical particle counter (OPC)/differential mobility particle sizer (DMA)/ AMETYST	size distribution of total and non-volatile aerosol 4 - 2 µm, absorption	Sauer, DLR	Voigt et al. (2017)
Ultra High Sensitive Aerosol Sizer (UHSAS)	Number size distribution of aerosol particles in a size range from 60 nm to 1000 nm	Pöhlker, Pöschl, MPIC	Andreae et al. (2018); Mei et al. (2020)
Aircraft-based Laser ABlation Aerosol MAss Spectrometer (ALABAMA)	Chemical composition of aerosol/cloud residual $\gtrsim 200\mathrm{nm}$	Schneider, MPI-C	Brands et al. (2011); Clemen et al. (2020)
Soot particle mass and absorption measurement system (SOPAMA)	rBC single particle masses and bulk aerosol absorption coefficients at 3 wavelengths	Sauer, DLR	
Backscatter Cloud Probe with Depolarisation (BCPD)	cloud particles 2 to 42 µm, aspherical fraction	Jurkat-Witschas, Lucke, DLR	Lucke et al. (2023)
spectrometer of the Munich Aerosol Cloud Scanner (specMACS)	spectral radiance (400-2500nm, 32.7° to 35.5° field of view), Stokes vectors (91°×117° field of view, RGB color channels)	Mayer, LMU	Ewald et al. (2016); Weber et al. (2024)
Alrborne H2O Mass Spectrometer (AIMS)	H_2O , $0.1 - 400 \text{ ppmv}$	Kaufmann, DLR	Kaufmann (2016)
Lyman Alpha Fluorescence Hygrometer (FISH)	H ₂ O, total water or gas phase water 1-1000 ppmv	Krämer, FZJ	Krämer (2009)
Sophisticated Hygrometer for Atmopheric ResearCh (SHARC)	Tunable Diode Laser Hygrometer, gas phase water vapor 2-40000 ppmv	Zöger, DLR	Kaufmann et al. (2018)
Water Vapor Analyser (WARAN)	Tunable Diode Laser Hygrometer, total water or gas phase water 1-40000 ppmv	Marsing, Voigt, DLR	Voigt et al. (2017) Marsing et al. (2023)
Fast ozone analyzer (FAIRO)	O ₃ , mixing ratio	Zahn, KIT	Zahn et al. (2012)
AtmosphEric Nitrogen oxides MEAsurement System chemiluminescence detector (AENEAS)	NO, NO ₂ , 10 pptv - 60 ppbv	Ziereis, DLR	Ziereis et al. (2022)
BAsic HAlo Measurement And sensor System (BAHAMAS)	T, p, u, v,meteorological and aircraft state parameters	Giez, DLR	Giez et al. (2021)

5. Forecast products and flight planning

A set of tailored forecast products was developed for CIRRUS-HL with the aim to visualize predictions of natural and aircraft-induced cirrus occurrence and their thermodynamic environment, to estimate cirrus properties in relation to distinct formation pathways, and to support flight planning. To illustrate the range of utilized flight planning tools, Fig. 3 shows selected products for planning the flight on 12 July 2021. The flight planning process involved the analysis of routinely produced charts at different pressure levels and forecast lead times. The detailed elaboration of flight plans was supported by interactive visualizations (Fig. 3a and b) using the Mission Support System (MSS; Rautenhaus et al. 2012; Bauer et al. 2022), e.g., to create vertical sections showing ECMWF forecast data along planned flight tracks. Fig. 3a gives an overview of the synoptic situation and the cirrus regions at different latitudes covered by the flight from Oberpfaffenhofen to Iceland. As dedicated cloud type sampling was pursued and different cloud formation processes were investigated, we employed the Lagrangian microphysics model ClaMS-Ice, which calculates detailed cirrus microphysical properties along 3D trajectories from ECMWF by means of a twomoment ice microphysics scheme (Luebke et al. 2016; Baumgartner et al. 2022). CLaMS-Ice products are: IWC (see Fig. 3b), ice crystal number concentration $N_{\rm ice}$, ice crystal mean mass size $R_{\rm ice}$, origin classification of cirrus (liquid or in-situ), and the identification of the ice nucleation mechanism (heterogeneous or homogeneous).

Related to a low pressure system west of Norway, an in-situ cirrus (light blue) region was predicted east of Iceland (Fig. 3a and e). An additional band of mostly liquid-origin cirrus (dark blue) over Iceland was also predicted by backward trajectories using the offline tool (LAGRANTO; Wernli and Davies 1997; Sprenger and Wernli 2015) and wind fields from ECMWF forecasts (Fig. 3d). Contrail optical thickness (Fig. 3c), calculated by the Contrail and Cirrus Prediction tool Co-CiP model (Schumann 2012) with air traffic data from previous years, predicted contrail clusters embedded in cirrus over Germany and north of England. In addition to the classic 2D forecast products, the interactive visual analysis software "Met.3D" (Rautenhaus et al. 2015; Schäfler and Rautenhaus 2023) was used for rapid 3D exploration of available forecast data (as done before, e.g., during the NAWDEX campaign (Schäfler et al. 2018)). 3D visualizations included, e.g., displays of forecast cloud fields (Fig. 3f); a detailed description of our experiences using 3D visualization during CIRRUS-HL was described by Schäfler and Rautenhaus (2023).

In preparation for the campaign, we also applied the global aerosol-climate model EMAC (ECHAM/MESSy Atmospheric Chemistry model; Jöckel et al., 2010) including the MESSy (Modular Earth Submodel System) aerosol microphysics submodel MADE3 (Modal Aerosol Dynamics model for Europe, adapted for global applications, third generation; Kaiser et al., 2014; 2019; Beer et al., 2020) coupled to (cirrus) clouds via a two-moment cloud-scheme (Kuebbeler et al. 2014; Righi et al. 2020). The simulation data used for the flight planning are based on the experiments BASE and \$14F01 of Righi et al. (2021). Climatological means were calculated from the 15-year (2001-2015) simulations to characterize the atmospheric distribution of potential INPs (e.g. aviation soot) and of ice crystals nucleated via heterogeneous and homogeneous freezing, in order to identify regions where INP-cirrus interactions could potentially be observed during the campaign. Fig. 3g shows the model climatological distribution of aviation soot particle number concentration in the upper troposphere in summer, while Fig. 3h and i depict upper-tropospheric number concentrations of ice crystals nucleated via heterogeneous freezing on aviation soot particles and via homogeneous freezing, respectively. The climatological distributions were used during flight planning to target cirrus regions potentially affected by aviation emissions and homogeneous nucleation. In a separate study, we will compare the measured occurrence and properties of aerosol and cirrus clouds with the model climatology to assess representativity.

6. Overview of cloud, aerosol and trace gas distribution during CIRRUS-HL

In this section, we present an overview of ice microphysical properties, trace gases and aerosol distributions measured during CIRRUS-HL focusing on the contrast between mid and high latitudes in June and July 2021.

a. Microphysical properties of cirrus at mid and high latitudes

One of the central aims of CIRRUS-HL was to investigate and contrast mid- and high-latitude microphysical cirrus properties. We here report on two data sets from two different combination probes, the cloud combination probe (CCP) mounted on the outbound right wing position and the NIXE-CAPS mounted on the central left wing position. The occurrence frequencies of ice water content (IWC), ice crystal number concentration (N_{ice}) and size (mean mass radius R_{ice} or ED) are shown in Fig. 4. In addition, the median, the 10th and 90th percentiles of the climatology

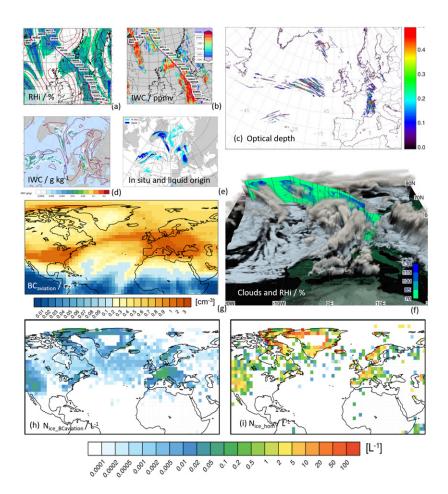


Fig. 3. Selected forecast products used for the planning of the flight on 12 July 2021: (a) MSS plot showing the planned flight track on top of RHi (color, %) and geopotential height (contours, m) at 250 hPa of the ECMWF HRES forecast from 00Z 12 July 2021, valid at 12Z 12 July 2021, (b) forecast of IWC from ClaMS-Ice trajectories (color, ppmv), (c) CoCiP forecast of contrail optical thickness (color), (d) forecast of IWC of liquid-origin cirrus based on LAGRANTO trajectories (color, g/kg), (e) forecast of liquid-origin (light blue) and in-situ (dark blue) cirrus from ClaMS-Ice, (f) Met.3D volumetric visualization of forecast clouds, together with a vertical section along the flight track showing RHi (same colorbar as in (a)), and climatological distributions in June and July at 260 hPa from the global model EMAC with MADE3 microphysics of (g) black carbon number concentrations from aviation and of number concentrations of ice crystals nucleated via (h) heterogeneous freezing on aviation soot particles and (i) homogeneous freezing.

from Krämer et al. (2020) for 1K-temperature bins are given. A summary of the microphysical properties of cirrus clouds from all flights from the CCP is given in Fig. 4a, b and c, while the

data from the NIXE-CAPS separated for mid and high latitudes are shown in panels d to f and g to i, respectively. In general, the probes showed a good agreement throughout the mission. The IWC (panel a) increases with increasing temperature, while N_{ice} (panel b) has no clear trend, since the increase in IWC is caused by larger ice particles (see panel c, ED) that have grown due to a higher amount of available water vapour in the warmer part of the cirrus temperature range. To get an impression of the types of cirrus observed in high and mid latitudes over Europe during CIRRUS-HL, we examine the IWC in more detail (panels a, d, g) relative to the median, the core minimum and the core maximum IWC from Schiller et al. (2008). According to the study by (liquid origin-cirrus, Krämer et al. 2016), combing cirrus simulations with observed climatologies, IWCs above the median and even above the core maximum line originate from glaciated mixed-phase clouds formed at lower altitudes that are uplifted in strong convective updrafts over a great vertical distance and thus carry a large amount of frozen water. Vertical velocities were measured by HALO. In addition, updraft speeds along the backward trajectories were analyzed. Under these atmospheric conditions of high updraft velocities which were encountered twice during dedicated convective flight above the Alps (Schäfler and Rautenhaus 2023), the IWCs reached values up to \approx 3 g m⁻³ (\approx 2000 ppmv, see also Supplement) that also led to aircraft pitot icing (Kalinka et al. 2023).

These thick cirrus occurred mainly in mid latitudes at higher temperatures and lower altitudes (\gtrsim 220 K, \lesssim 10 km, Fig. 4d). Based on classification from Krämer et al. (2016, 2020), observations between the median and the core maximum IWC can be assigned to liquid origin-cirrus in frontal systems, where the updrafts are less intense than those in convective cirrus. Using this classification, approximately half of the cirrus probed in mid latitudes during CIRRUS-HL were liquid origin-cirrus (see Fig. 1 and (De La Torre Castro et al. 2023)). The thinner cirrus with low IWCs mostly appeared on top of frontal systems or in high pressure systems. N_{ice} shows overall occurrence frequencies slightly above the median in the mid latitudes (Fig. 4e), indicating contrail cirrus or cirrus formed in high updrafts. The occurrence frequencies of mid-latitude ice crystal sizes (R_{ice}) are shown in Fig. 4f. The smallest R_{ice} have been observed together with the highest N_{ice} around 212-216 K. These observations can be traced back to young contrails and contrail cirrus using coincident measurements of nitrogen oxides and non-volatile particulate matter.

In the Arctic, greater variability is observed in the cirrus IWC compared to the mid-latitude cirrus (Fig. 4g). Based on backward trajectory analysis, 30% thin in situ-cirrus with low IWC and 70% thick liquid origin-cirrus with higher IWC, mostly advected from the mid latitudes into the Arctic, were probed. This indicates a significant influence of mid-latitude air on the Arctic cirrus composition. The enhanced frequencies of very low IWCs ($\lesssim 1$ ppmv) correspond to also very low N_{ice} of large sizes (Fig. 4h and i) indicating cirrus formed in high latitudes and reflecting calm conditions and low concentrations of INPs at high latitudes.

In summary, during CIRRUS-HL a wide variety of atmospheric conditions and cirrus with different microphysical properties were sampled, ranging from very thick convective liquid origin to very thin in situ origin cirrus and contrail cirrus. We found larger ice crystals associated with an order of magnitude lower ice crystal number concentrations in the Arctic than at mid latitudes (De La Torre Castro et al. 2023). This is potentially related to the combination of low INP concentrations and low updrafts at high latitudes and the higher amount of air traffic and convection at mid latitudes.

b. Aerosol and trace gas distribution in high and mid latitudes

Flight strategies and targeted altitudes during CIRRUS-HL focused not only on the cirrus altitudes but also on the full vertical distribution of aerosol and trace gases, scanning the atmosphere from the boundary layer to the stratosphere. Fig. 5 shows altitude profiles of median aerosol and trace gas distributions, with 25th and 75th percentiles of the distribution in equidistant altitude bins. Aerosol number concentrations (Fig. 5 a and b) were measured using a condensation particle counter (CPC) of the AMETYST instrument with a lower cut-off diameter of 12 nm. In Fig. 5a, we compare the mid-latitude profile with measurements during the COVID-19 lock-down period during the BLUESKY mission (Voigt et al. 2022) and a ten-year data set of aerosol measurements in June and July for the region covered by CIRRUS-HL in pre-COVID times from the CARIBIC project (Zahn et al. 2024). As CIRRUS-HL took place during a period of increasing anthropogenic activity including air traffic, mean aerosol number concentrations in the free troposphere are mostly above the BLUESKY measurements but still reduced 40 to 80 % compared to the CARIBIC dataset. Mean near-surface (< 3 km) levels of aerosol particles (Fig. 5 a and b) as well as of combustion related trace gases such as CO and NO (Fig. 5c and d) were enhanced in mid latitudes compared to

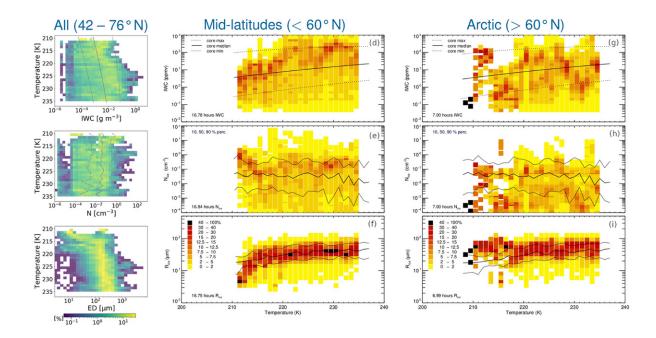


Fig. 4. Frequencies of occurrence of the cirrus microphysical properties (IWC, N_{ice} , ED, R_{ice}) with respect to temperature. Left column (panels a, b, c): all measurements $38 - 76^{\circ}N$ (from CCP + PIP, De La Torre Castro et al. (2023)); middle and right columns (panels d, e, f and g, h, i): mid-latitude (> $60^{\circ}N$) and Arctic (< $60^{\circ}N$) measurements (from NIXE-CAPS, Krämer et al. (2016)). IWC: Ice water content, calculated from the ice particle size distributions by using the mass-dimension relation shown in Krämer et al. (2016); Afchine et al. (2018), N_{ice} : ice crystal number concentration, R_{ice} : ice crystal mean mass radius (~ IWC/ N_{ice}), ED: ice crystal effective diameter; IWC lines: minimum, median and maximum of the IWC core range from Schiller et al. (2008); N_{ice} , R_{ice} lines: 10, 50, 90th percentiles from Krämer et al. (2020).

high latitudes, related to the greater anthropogenic activity in mid latitudes. In the free troposphere (between 6 and 10 km), this trend is inverted, potentially due to cloud scavenging effects on black carbon and other aerosol particles (Zieger et al. 2023). Further, LAGRANTO backward-trajectory calculations indicate that long-range transport of biomass burning emissions from Siberia and North America led to elevated aerosol and CO levels during some events, exceeding 200 ppbv in the upper Arctic troposphere. One case reaching the stratosphere, measured on 7 July 2021, is shown in Fig. 8. In flight corridors, the aerosol concentrations are higher in the mid latitudes. The NO concentration profile (Fig. 5d) generally shows higher median values in mid latitudes throughout the troposphere and particularly enhanced at cruise altitudes. Given its short lifetime, elevated NO concentrations indicate fresh emissions. Main sources of NO in the upper troposphere

are transport from the polluted boundary layer and in situ production by lightning (Schumann and Huntrieser 2007) and air traffic (Jurkat et al. 2011). All sources are more pronounced at mid latitudes than at higher latitudes. At the altitude of cirrus formation, we find a significantly smaller difference in mean total aerosol number concentration from high to mid latitudes compared to the mean ice crystal number concentration (almost one order of magnitude). Future studies will concentrate on the distribution of INPs to assess the impact on cirrus cloud formation at high and mid latitudes.

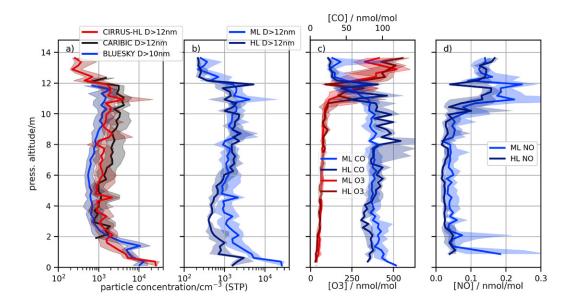


Fig. 5. Altitude profiles of aerosol and trace gases for high and mid latitudes. (a) median mid-latitude aerosol number concentration profiles from CIRRUS-HL (D>12nm), BLUESKY (D> 10nm) (Voigt et al. 2022) and CARIBIC (D> 12nm) (Zahn et al. 2024), where D is the lower cut-off diameter of the CPC measurement. (b) CIRRUS-HL high- and mid-latitude aerosol profiles of total aerosol number concentrations at standard conditions (STP). (c) Median FAIRO O₃ (red) and Picarro CO (blue) mole fractions. (d) Median AENEAS NO mole fractions. In panels (b)-(d) light colors represent mid-latitude measurements (<60°N), darker colors high-latitude measurements (>60°N). In all panels shaded areas indicate the range between the 25th and 75th percentiles, solid lines refer to the median.

c. Water vapor distribution in high and mid-latitudes

Upper tropospheric humidity is a key quantity for cirrus formation and life cycle (Diao et al. 2015) and governs the formation, persistence and microphysical properties of contrails and their

evolution into contrail cirrus (Schumann et al. 2015; Verma and Burkhardt 2022; Wang et al. 2025). We provide in Fig. 6a a distribution of the frequency of occurrence of relative humidity with respect to ice (RHi) at temperatures below 235 K from all humidity sensors aboard HALO during CIRRUS-HL. The distributions are given for measurements inside of clouds from SHARC and clear-sky conditions from WALES, FISH, AIMS and SHARC (for H₂O < 80 ppmv in light colors). The instruments and derived RHi from temperature measurements aboard HALO (Giez et al. 2021) and ECMWF temperature fields agree well within their respective measurement and uncertainty range (Kaufmann et al. 2018), providing a clear picture of the humidity for ambient temperatures below 235 K. While each instrument has an uncertainty of less than 10% for the measurement of water vapor concentrations, the uncertainty of the derived RHi is on the order of 10 to 12% due to the additional uncertainty of the temperature measurement (Kaufmann et al. 2018). The summer upper troposphere during CIRRUS-HL exhibits a high occurrence frequency of supersaturated air, both inside the clouds as well as in clear-sky conditions. Inside the clouds, a median RHi of 107% is found, with 25th and 75th percentiles of 99 and 140%, respectively. From the FISH and SHARC measurements it is found that 8% and 14% of the clear-sky data are supersaturated, respectively. We compare the SHARC RHi frequency distributions in liquid-origin and in situ origin cirrus next to contrail cirrus for high and mid latitudes. Medians (indicated by dashed vertical lines) of RHi distributions in all naturally formed cirrus types are above saturation. The highest values of in-cloud supersaturation have been found in the high latitudes inside in situ origin cirrus. These thin cirrus with low ice crystal numbers were mostly formed in the pristine Arctic regions with a reduced number of INPs, potentially related to the lower levels of pollution (Ovarlez et al. 2002; Jensen et al. 2001). During CIRRUS-HL, contrails and contrail cirrus are often detected in regions with high RHi inside or at the top of cirrus (see Fig. 9) (Jensen et al. 1998; Dekoutsidis et al. 2023). These observations are in contrast to previous measurements in spring (Voigt et al. 2017) where contrail cirrus were mainly observed in subsaturated conditions (Li et al. 2022). We explain the systematically supersaturated conditions (median values above 100%) observed in mid latitudes with two flights in convectively-forced cirrus during summer which due to higher updrafts may have altered the RHi spectrum to higher supersaturation (Petzold et al. 2017; D'Alessandro et al. 2017). During transfer from Oberpfaffenhofen to higher latitudes, we frequently flew at cloud tops, some of them convectively-forced, where enhanced supersaturation

is often observed (Diao et al. 2014a,b; Dekoutsidis et al. 2023). In high latitudes, the reduced number of INPs and the thin cirrus of low number concentrations (Krämer 2009; Patnaude et al. 2021) and surface area do not provide a large condensational sink to relax RHi to 100%. The median RHi in contrail cirrus are shown in black in Fig. 6b and c. Due to the typically high number concentration of ice crystals ($N_{ice} > 0.1 \text{ cm}^{-3}$) with low effective diameters (ED< 40 μ m), the RHi median relaxes towards saturation, as previously observed in young contrails (Kaufmann et al. 2014). This observation indicates the relevance of contrail cirrus as an effective sink of ambient supersaturation, thereby suppressing new particle formation and particle growth (Schumann et al. 2015).

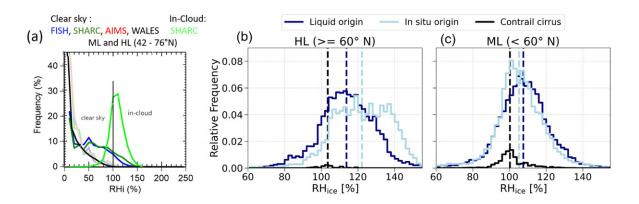


Fig. 6. Overview of RHi measurements during CIRRUS-HL. (a) Frequency distributions for mid and high latitudes and temperatures $\lesssim 235$ K (based on more than 30000 data points for each distribution): (i) clear sky conditions from SHARC and FISH and selected for upper tropospheric values ($H_2O<80$ ppmv) for SHARC, FISH, AIMS and WALES; (ii) in-cloud conditions from SHARC. (b) and (c) In-cloud relative frequency distribution from SHARC, separated into three cloud types: in-situ (light blue), liquid-origin (navy) and contrail cirrus (black) for high (b) and mid-latitude (c) cirrus. Medians of contrail cirrus, liquid-origin and in situ origin-cirrus range from 100% (ML contrail cirrus) to 105% (ML in-situ) and 122% (HL in-situ), and are marked with dashed lines.

7. Selected results from the CIRRUS-HL mission

This section provides selected results from individual flights during CIRRUS-HL. The combination of measurements and models from these flights give short insights into the flight strategies, the regions and the multitude of applications and research topics that CIRRUS-HL targeted. We first focus on one case study on natural Arctic clouds, on aerosol and ice particle residual properties and radiation measurements. The last two selected results focus on contrail cirrus measurements and modelling.

a. Impact of a high latitude cirrus on the radiation budget

To quantify the impact of cirrus on the Arctic radiative budget, ice crystal optical properties and spectral albedo were assessed during CIRRUS-HL. On 29 June 2021, a staircase flight pattern analyzed a high-level warm front cirrus north of Iceland, consisting of flight legs below, within, and above it. Fig. 7a shows the predicted ice and liquid water content across the HALO flight path from the ECMWF's 12 UTC analysis. Flight leg 5, the system's center, recorded the highest LWP (Fig. 7b).

The impact of the clouds on the spectral albedo measured by SMART is indicated in Fig. 7d for each flight leg. Two dependencies are obvious. The albedo in the visible spectrum (e.g., around 500 nm wavelength) increases with increasing amount of liquid and ice water below HALO as calculated from the model. This spectral range also dominates the broadband albedo. The lowest albedo is found in flight leg 1 while the maximum albedo is observed in flight leg 5. Analyzing the spectral albedo in the near infrared region between 1500nm and 1750nm, where absorption of the ice and liquid particles is apparent, allows for characterizing the impact of cloud thermodynamic phase (Ehrlich et al. 2008). Flight leg 1 and 2 show a high albedo in this spectral range and, thus, the albedo is still mostly influenced by the lower level liquid cloud. Starting with flight leg 3, the impact of the ice cloud on the spectral albedo becomes apparent. Fig. 7c shows the asymmetry parameter g retrieved from the PHIPS measurements using the methodology presented in Xu et al. (2022) and its standard deviation along the flight track. A total of 1,716 crystals were sampled, and one retrieved g value corresponds to a population of 50 crystals (about 19 L volume). Measured g values ranged from 0.69 to 0.76, averaging 0.72, indicating the presence of complex crystals. This is in agreement with the high-resolution microscopic single particle images (Fig. 7e). Despite the variety in crystal shapes across different flight legs — with flight leg 2 dominated by 72% side planes, flight leg 3 showing 54% irregular polycrystals, and higher altitudes featuring over 70% bullet rosettes — g showed minimal variation. This finding contradicts higher g values typically used in radiative transfer simulations, highlighting the potential

underestimation of cirrus cloud reflectivity. Therefore, direct measurements of ice crystal single scattering properties in combination with spectral albedo measurements can be used to assess the current representation by ice optical parameterizations in numerical models and eventually to develop new optical parameterizations.

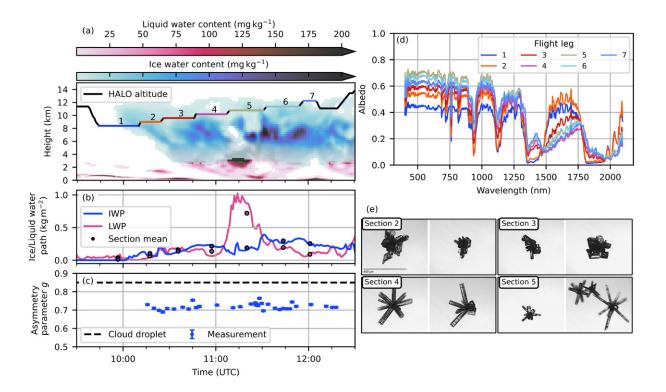


Fig. 7. Investigation of an Arctic cloud. (a) Staircase flight pattern from F05 on 29 June 2021 shown together with a cross section of IWC and LWC from the 12 UTC analysis of the ECMWF's IFS. (b) IWP and LWP along track with the section mean from the IFS's 12 UTC analysis. (c) Mean asymmetry parameter *g* along the flight track. (d) Mean spectral albedo for the seven staircase sections. (e) Collage of ice crystal images from different sections.

b. Measurements of cirrus particle residuals, upper tropospheric fine mode aerosol particles, and their ice nucleation ability

Selected results from research flights F08 to F11 representing different atmospheric conditions are presented here: Lower stratospheric (LS) biomass burning (BB) aerosol (*LS_BB_aerosol*, F10/11) identified through chemical single particle composition analysis, LS aerosol (*LS_aerosol*,

F08/09), upper tropospheric (UT) aerosol (*UT_aerosol*, F10/11), and UT cirrus particle residuals (*UT_residuals*, F10/11). According to backward trajectory analysis and satellite images, the *LS_BB_aerosol* likely originated from wildfires in British Columbia, Canada, where pyrocumulonimbus clouds reached the UTLS seven days prior to our measurements.

As illustrated in Figure 8a, the background concentrations of large particles (≥ 500 nm) were similarly low for *UT_aerosol* (green) as for *LS_aerosol* (grey), but the concentration of Aitken mode particles ($\leq 100 \,\mathrm{nm}$) was much higher for $UT_aerosol$, which may be due to UT aerosol formation (e.g., Brock et al. 1995). As the LS_BB_aerosol (orange) exhibited no enhanced Aitken mode but markedly higher concentrations of accumulation mode particles (≥ 200 nm), we suggest that BB smoke had no major impact on the Aitken mode aerosol in the LS. Similar to the UT_aerosol concentration, the *UT_residuals* (blue) also show a maximum in the Aitken mode size range, in line with earlier size distribution measurements of cirrus particle residuals (Seifert et al. 2003). This feature is also commonly found for ice particle residuals in mixed-phase clouds (Mertes et al. 2007; Kupiszewski et al. 2016), which are formed by heterogeneous ice nucleation, suggesting that this mechanism was observed in the investigated cirrus clouds, too. Figure 8b shows the particle groups resulting from the mass spectrometric analysis. For LS_aerosol, the particle group "meteoric material" (e.g., Murphy et al. 1998; Schneider et al. 2021) was by far the most frequently observed. Furthermore, in comparison to the other sampling periods, a higher fraction of the particle group "elemental carbon" and, in comparison to *UT_aerosol*, a higher fraction of the particle group "organic type 2" was observed. For LS_BB_aerosol, mainly particles indicating BB were identified. For these samples a higher fraction of mineral dust and sea salt particles compared to LS_aerosol was observed. These particles may have been lifted to the UTLS within the pyrocumulonimbus (e.g., Andreae et al. 2004). *UT_aerosol* is used in direct comparison to *UT_residuals* since both measurements took place during F10/11 under similar conditions (altitude, temperature, ozone, time of day, and location). The composition of the *UT_residuals* is dominated by "organic type 3", sea salt, and mineral dust, which together account for about 80 % of the particles. In contrast, these three particle groups account for less than 4% of the UT_aerosol, indicating that heterogeneous ice nucleation was important in the formation of the cirrus cloud particles sampled during F10/11. Fig. 8c shows the temperature dependent INP concentrations derived from HERA for filters sampled during LS_BB_aerosol, LS_aerosol, and UT_residuals under similar conditions. The

LS_aerosol filter can be regarded as representative for the background INP concentrations prevailing in the LS. Looking at Fig. 8c it becomes obvious, that LS background INP concentrations (STP) were significantly lower than typical mid-latitude tropospheric INP concentrations, as derived from precipitation samples (grey area; Petters and Wright 2015). Astonishingly, INP concentrations in the LS_BB_aerosol were found to be lower than those representative for the LS background, i.e., seemingly, LS_BB_aerosol particles are not significantly contributing to the LS INP population. To allow for direct comparison, the number of INPs per filter was normalized with the total number of aerosol particles or cirrus particle residuals per filter retrieved from the FASD- and CVI-UHSAS, respectively (Fig. 8d). The *UT_residuals* featured a higher relative number of INPs compared to LS_aerosol and LS_BB_aerosol. Nonetheless, at -30 °C the fraction of immersion freezing INPs vs the total number of residuals was only slightly above 0.02 \%, suggesting that immersion freezing above -30°C only played a minor role in forming ice crystals in these cirrus clouds. Either the ice crystals are formed by other, potentially more numerous immersion INP below -30°C or via other pathways such as homogeneous freezing, secondary ice production, etc.. The low fraction of INP in the LS_BB_aerosol suggests an insignificant contribution of BB aerosol particles to the INP population.

c. Remote sensing of contrails and contrail cirrus above Germany

Contrail and contrail cirrus predicted by the CoCiP model were targeted in CIRRUS-HL flights above Europe. One case of contrails and related fall streaks was observed by remote sensing instrumentation on 25 June 2021. The coincident measurements of backscatter ratio and particle depolarization at 532.1 nm from the HSRL Lidar WALES shown in Fig. 9d and e show vertical cross sections of two mixing cloud systems with different origins represented by a high and low depolarization ratio (Fig. 9e) (Urbanek et al. 2018). The first section of the cloud (before 14:22 UTC) was influenced by a southerly flow, while the second part had a more pristine character with air masses coming from Northern latitudes. Above upper level cloud tops, water reached saturation with respect to ice also measured by WALES (Fig. 9f between 14:16-14:24 UTC) (Groß et al. 2014) which allowed formation of persistent contrails. The high backscatter ratios (Fig. 9e) at the top of the cloud indicate contrails and related fall streaks (Sassen 1997; Jensen et al. 1998; Unterstrasser et al. 2017) with ages of several minutes extending vertically several hundred meters below the

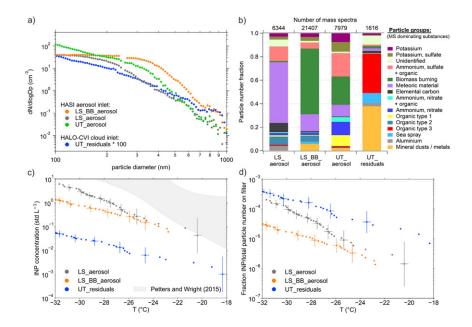


Fig. 8. Physical and chemical characteristics of aerosol particles and cirrus particle residuals under different atmospheric conditions, i.e., lower stratospheric biomass burning aerosol (LS_BB_aerosol, F10/11, averaged ozone concentration and altitude during measurement: 474 ppb and 13317 m), lower stratospheric aerosol (LS_aerosol, F08/09, 450 ppb and 13150 m), upper tropospheric aerosol (UT_aerosol, F10/11, 83 ppb and 10564 m), each sampled through the HASI inlet, and upper tropospheric cirrus particle residuals (UT_residuals, F10/11, 81 ppb and 10406 m) sampled through the HALO-CVI. Averaged ambient air temperature ranged between 223 and 227 K during the four periods. a) Aerosol particle size distributions during the time periods described above measured with the FASD-UHSAS and cirrus particle residual size distribution measured with the CVI-UHSAS. The size distribution for the cirrus particle residuals is scaled by a factor of 100 for better comparison. b) Composition of the aerosol particle and the cirrus particle residual population determined with the ALABAMA. Particles are sorted into groups according to those ion peaks that dominated the individual mass spectra. c) INP concentrations per liter of sampled air from HERA filters as a function of freezing temperature. The grey area in the background shows typical tropospheric INP concentrations from mid-latitudes (Petters and Wright 2015). Sampling volumes were calculated for standard conditions (T=273.15 K, p=1013.25 mbar), and in the case of the CVI filter normalized with the average enrichment factor of the sampling period. d) Temperature dependent INP fraction (INP number divided by total number of aerosol particles or cirrus residual particles sampled on HERA filters). Aerosol particle concentrations were taken from the FASD-UHSAS, cirrus residual particle concentrations from the CVI-UHSAS.

flight altitude into the cirrus deck. In the upper, dense part of the contrail no particle depolarization could be measured due to a saturated signal. Previous measurements have shown that contrail to cirrus evolution causes a change in the scattering phase functions and the asymmetry parameter g (Gayet et al. 2012; Chauvigné et al. 2018). The sedimenting contrail ice crystals locally modify the particle depolarization of the cloud below. This change in depolarization from the contrails has been observed simultaneously by the downward-looking hyperspectral and polarized imager specMACS (Ewald et al. 2016; Weber et al. 2024). Fig. 9a, b show the degree of linear polarization and the corresponding RGB image measured by the polarization cameras. The images highlight an optically thin line-shaped contrail oriented in across-track direction, which corresponds to the contrail measured by WALES at 14:20 UTC. An optically thin cirrus and a layer of small cumulus clouds are visible below. The contrails have a smaller degree of linear polarization and appear darker in Fig. 9a. Furthermore, the cloudglint which is formed by specular reflection on oriented ice crystals is visible as a narrow bright spot in the upper left corner, and indicates the presence of oriented ice crystals in the cirrus. Moreover, the cloudbow which forms by scattering on liquid cloud droplets in the lower cumulus layer appears as a bow of enhanced polarization in the lower right and has been evaluated to determine the droplet size distribution (Pörtge et al. 2023). The various contrails measured by WALES are also visible as white thin lines along the across-track angle perspective in the observation of the hyperspectral camera of specMACS at 1390 nm in Fig. 9c. These stripes co-incide with the higher backscatter ratio of WALES. We chose a wavelength of 1390 nm with strong water vapor absorption so that, in contrast to the RGB image (visible wavelength range), only high clouds above the absorbing layers are visible.

The coincident measurements give a 3-D information of young contrails above a thin cirrus deck, providing evidence on the change in the polarization of the incident light due to the contrails and in contrast to the natural cirrus below.

d. Cloud Radiative Forcing as observed from satellite observations with CoCiP model studies

Satellite observations can give a general overview of contrail cirrus and natural cirrus conditions in a large spatial scale. Due to smaller crystal sizes, young contrails tend to show higher brightness temperature differences (BTD (10.8-12.0), channel 10.8 µm minus 12.0 µm) compared to natural cirrus (e.g. Mannstein et al. 1999). We conducted an analysis of optical properties and radiative

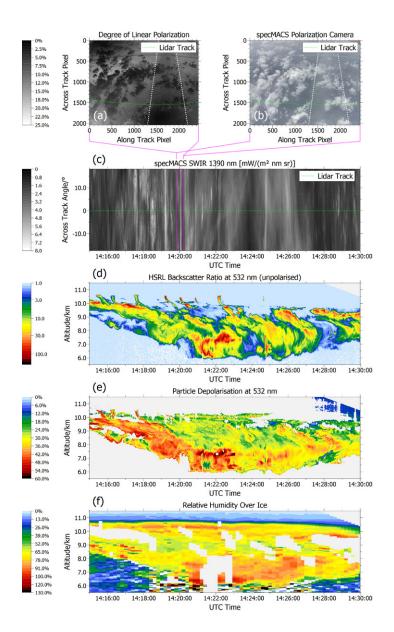


Fig. 9. 3D-observations of contrails formed at the upper level cloud tops. (a) Degree of linear polarization and (b) RGB image from specMACS polarization camera at 14:20 UTC. Dashed lines indicate position of a young contrail. (c) 2-D radiance measurement from specMACS at 1390 nm. (d) WALES backscatter ratio at 532 nm of young contrails and cirrus during one flight on 25 June 2021 observed above Germany. (e) Simultaneous measurement of particle depolarization at 532 nm from WALES. (f) Relative humidity over ice from WALES and ECMWF temperature.

impact of contrail cirrus based on Meteosat Second Generation (MSG) satellite observations and compared the results with CoCiP simulations at 10:00 UTC on 21 July 2021. CoCiP simulations

adopted a humidity correction methodology (Teoh et al. 2022a) to improve the input RHi derived from ECMWF data. This correction accounts for the dry bias observed in the upper troposphere (Teoh et al. 2022b; Gierens et al. 2020; Wang et al. 2025) and its consequential influence on the persistence and climate impact of contrail cirrus. In Fig. 10a, an outbreak of contrail cirrus, represented by white areas, is situated over low-level liquid clouds, indicated by yellow regions, spanning the North Atlantic Ocean and the Iberian Peninsula. The flight path of HALO marked with a yellow line reflects the occurrence of scattered ice clouds with diameters larger than 3 μm measured with the CCP (purple shading). The linear-shaped contrails are clearly depicted as white lines in the BTD image in Fig. 10b. Satellite observations in Fig. 10c processed with the CiPS (Cirrus Properties from SEVIRI, Strandgren et al. 2017) algorithm reveal a contrail optical thickness of approximately 0.35, similar to the results in Wang et al. (2023), which aligns closely with the corresponding CoCiP simulations displayed in Fig. 10d. However, CiPS is not able to detect all contrails that can be observed by eye in the RGB composite in panel b. Examining the radiative effects, contrails in Fig. 10e exhibit a positive net cloud RF of approximately 5-10 W m⁻² over the ocean surface, in contrast to a negative net RF of about -5 W m⁻² over northern Portugal and Spain, derived from satellite observation-based radiative transfer modeling as in Wang et al. (2023). Contrails forming above low-level clouds with a high albedo in this case over ocean are more likely to be warming because the incoming solar radiation would have been reflected by the low-level clouds regardless of the presence of contrails, thereby reducing the contrail SW RF (Teoh et al. 2022a). CoCiP simulations, as illustrated in Fig. 10f, also present the warming-cooling contrast in the radiative effects of contrail cirrus, with the net RF over the continent tending towards a more cooling effect. The reason for this can be attributed to the somewhat greater optical thickness and increased reflection observed in the CoCiP simulation, as shown in Fig. 10d. Future work will connect the airborne in situ and remote sensing data collected during flight to the satellite retrieval and model result presented here.

8. Summary and Outlook

The HALO mission CIRRUS-HL provides a new data set that includes a large spectrum of cloud, aerosol, trace gas, and radiation measurements in the mid-latitude dense air traffic regions and in pristine Arctic regions in June and July 2021. Differences between high-latitude and mid-

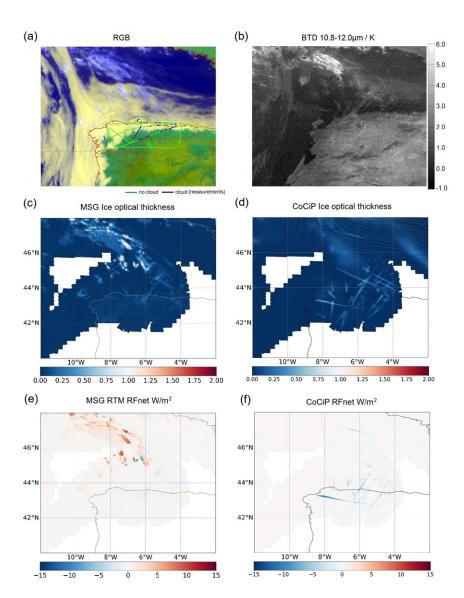


Fig. 10. Microphysical properties and radiative effects of a contrail cirrus outbreak over the North Atlantic Ocean, and northern Portugal and Spain on 21 July 2021. (a) RGB with a yellow line representing the HALO flight path and purple colors representing the contrail cirrus cloud flag from CCP for ice crystals larger than 3 µm. (b) BTD. (c) Contrail cirrus optical thickness from MSG observations. (e) Net RF through satellite observation-based radiative transfer modelling. (d) and (f) Equivalent parameters of (c) and (e) but simulated by the CoCiP model. Values in (c)-(f) are masked with CoCiP simulated cirrus grid boxes.

latitude cirrus, aerosol and trace gas distributions could be observed with influences from aviation, convectively formed cirrus, cirrus formed from the liquid phase (liquid-origin cirrus) and from the vapor phase (in situ cirrus). Differences in occurrence of supersaturation inside cirrus as well

as the abundance of total aerosol number showed contrasting patterns from the Arctic to the mid latitudes. We examined cases of Arctic cirrus and contrail cirrus formed at the cloud tops of natural cirrus and contrails cirrus above low level clouds to show the cloud radiative effect and their impact on depolarization.

As the present paper provides an overview of the wide variety of cirrus measurements preformed during CIRRUS-HL in different latitude ranges, the data set invites a more detailed analysis of the different targets. Planned studies will provide an overview on microphysical properties of contrail cirrus categorized by age and thermodynamic state as a basis for model intercomparisons in order to improve our understanding on the interaction and modification of contrail cirrus with clouds and to reduce uncertainties on contrail cirrus radiative forcing estimates and on contrail prediction. Aviation as a source for in situ aerosol containing a high fraction of "organic type 3" will be investigated based on the analysis of the composition of upper tropospheric aerosol residuals. The influence of INP type and number on the formation of cirrus clouds measured during CIRRUS-HL will be the focus of another study to contrast the influence of anthropogenic (e.g. air traffic) and natural sources in mid and high latitudes. The impact of the latitude dependence of cirrus on the linear depolarization ratio observed by the space borne lidar CALIOP aboard CALIPSO will give further insight on the differences of the formation pathways of these cirrus based on a multi annual data set. The sampling of two convective anvils during CIRRUS-HL offers the basis to investigate the impact on the local perturbation of LS water vapor through injection of ice crystals in overshooting tops of convective systems. To study the indirect aviation effect on high and low clouds, the follow-up mission AERO-CLOUD will concentrate on aerosol-cloud effects from aircraft emissions such as the impact of sulfate particles on low-level clouds and the impact of soot emissions on cirrus properties and ISSR characterisation. The data and results of CIRRUS-HL represent a valuable contribution to the international meteorological and aviation community through validation of weather forecast, contrail prediction tools and climate models.

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Data availability statement. Processed data from the CIRRUS-HL campaign will become publicly available on the HALO database (http://doi.org/10.17616/R39Q0T) at https://halo-db.pa.op.dlr.de/mission/125 in June 2026. Earlier access to the data is available by contacting the first author.

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