



Satellite-based evidence of regional and seasonal Arctic cooling by brighter and wetter clouds

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Abstract. Using two decades of satellite-based measurements of reflectance of solar radiation at the top-of-atmosphere and a complementary record of cloud properties, it is concluded that the loss of Arctic brightness due to sea ice retreat is not compensated by a pan-Arctic increase in cloudiness, but rather by a systematic change in the thermodynamic phase of cloud and a resultant effect on cloud reflectance. Liquid water content of the clouds has increased resulting in positive trends in susceptible cloud properties. Consequently, a cooling trend by clouds is superimposed on top of the pan-Arctic amplified warming, induced by the anthropogenic release of greenhouse gases, the ice albedo feedback and related effects. Except above the permanent and marginal sea ice zone around the Arctic circle, the rate of surface cooling by clouds has increased, both in spring (−32% in total radiative forcing for the whole Arctic) and in summer (−14%). The magnitude of this effect depends on both the underlying surface type and changes in the regional Arctic climate.

10 1 Introduction

The most recent climate projections indicate that the Arctic will be free of sea ice by the summer of 2035 (Guarino et al., 2020). The Arctic near-surface increase of temperatures is about twice the global average over the past four decades. This is referred to as “Arctic Amplification” (Serreze and Francis, 2006). The size of a temperature increase from a doubling of the carbon dioxide atmospheric loading was first quantified by Arrhenius in 1896 (Arrhenius, 1896). The impact of the anthropogenic release of greenhouse gases on the surface temperature has become an increasingly important topic of scientific interest, public debate and concern and international environmental policy, since at least 1990. However, the Arctic is a special case (Serreze and Barry, 2011).

Modeling the changing behavior of clouds sufficiently accurately is identified as the most uncertain factor in the climate projections of greenhouse gas forcing (Zelinka et al., 2020). This is in large part because clouds modulate the Arctic radiation budget in the shortwave (SW) and longwave (LW). Changes in the temperature, water vapor and the availability of cloud condensation nuclei result in turn in changes in the cloud absorption and reflectance. Thus, improved knowledge of the changing optical and radiative properties of clouds, the Earth’s surface and the resultant SW reflection and LW emission at the top of the atmosphere are essential to test and improve the accuracy of climate model projections. The most complex and ambitious measurement endeavours (Wendisch et al., 2019; Shupe et al., 2021), exploiting the synergy of on-ground, ship and airborne



25 techniques, rely on complementary measurements of satellite sensors which provide synoptic coverage of the Arctic clouds over long time scales.

Instruments aboard satellites measure the solar radiation scattered back to space from the Arctic surface and atmosphere constituents (i.e. ice, snow, ocean, land, clouds, trace gases and aerosols (Kokhanovsky and Tomasi, 2020; Serreze and Barry, 2014)). Each contribution has a different response to radiation depending on their physical properties. Incoming solar SW
30 radiation in the ultraviolet and visible is scattered strongly by ice, snow, and clouds, whereas open waters absorb significantly and scatter back to space much less electromagnetic radiation in the solar spectral range. On the other hand, LW radiation fluxes are also modulated by clouds, which may warm or cool both the Top-Of-Atmosphere (TOA) and the surface. The changes at the surface resulting from the interplay of changes in sea ice properties, snow and cloud properties in the atmosphere lead to a nonlinear response of the radiation budget in the Arctic to changes in temperature (Lindsay and Zhang, 2005).

35 Cloud fractional cover (CFC) over the Arctic can be as large 70% throughout the year (Karlsson and Devasthale, 2018). The magnitude and variability of CFC depends on atmospheric conditions, which impact on cloud nucleation and growth, and on the type of sensors used to measure it (Chan and Comiso, 2013). The CFC seasonal cycle exhibits a strong bimodal distribution, with a maximum up to 90% in summer along the North Atlantic throughout the circumpolar ocean waters and a minimum of 40% in winter months (Eastman and Warren, 2010b, a; Boccolari and Parmiggiani, 2018). Rather than having a
40 latitudinal dependence, CFC in the Arctic appears to be dependent on the underlying surface type, meteorology and topography. Distinct patterns having divergent CFC trends and types in the Arctic have been identified, which follow sea ice and open waters contours. Using the same data sets does not guarantee consistent analysis and interpretation of the observations. This is the case for Boccolari and Parmiggiani (2018), where CFC data, derived from AVHRR (see Tab. A1 for the meaning of all acronyms) over the Arctic between 1982 and 2009, is in unexpected and large disagreement with results from Schweiger (2004) and Wang
45 and Key (2005b), even though all three research groups use the same data.

Clouds are the most important atmospheric factor in the modulation of energy flow exchange between the Arctic and its surroundings. Sledd and L'Ecuyer (2019) partitions the relative importance of surface and atmosphere with respect to the changes of albedo at TOA. While the majority of the variability is controlled by surface reflection, TOA albedos are consistently influenced by radiative transfer in the atmosphere: the contribution of the atmospheric reflection being approximately 84% of
50 the total Arctic albedo. This finding is relevant for a melting cryosphere, in which the relative changes in surface reflection are offset by atmospheric reflection, primarily by the reflectance of clouds (Donohoe and Battisti, 2011). Consequently and as expected, the presence of clouds limits the impact of the changes of the surface reflectance on the spectral reflectance at TOA (R_{λ}^{TOA}). Hence, a decrease in summer CFC over Greenland is held responsible for the ice mass loss acceleration and, consequently, for the decrease in albedo. A decrease in cloudiness implies an increase of SW downwelling fluxes at the surface.
55 This is correlated with the synoptic North Atlantic Oscillation (Hofer et al., 2017) and anticyclonic activity promoting adiabatic tropospheric warming of subsiding air masses (Shahi et al., 2020).

In Pistone et al. (2014) a downward trend of all-sky albedo across the Arctic is reported. This is not compensated by an opposite trend in cloudiness, thus levelling out the recent pan-Arctic reflectance trend. However, this analysis is limited to oceanic regions and additional uncertainties are caused by the conversion from clear-sky to all-sky albedo at the beginning of



60 their record. As the clear-sky signal is derived from the sea-ice record with sensors for which the atmosphere is almost entirely transparent, the all-sky albedo is computed with a post-hoc method adding the atmospheric part and is not the outcome of direct satellite measurements. He et al. (2019) reports significant regional covariance of CFC and sea ice. Strong negative correlations are seen over Beaufort and East Siberian Seas. The highest CFC occurs typically over marginal sea ice regions. The latter is negatively correlated with sea ice anomalies. The magnitude of the surface albedo feedback is limited by the formation of
65 low-level clouds from the evaporation of the melted sea ice regions. In such conditions, clouds have a negative feedback, in the SW radiation spectral region. This offsets LW radiation emission. Overall, He et al. (2019) consider that CFC over the melted water influences the albedo change in the albedo feedback process. The extra CFC resulting from sea ice loss is a second-order contribution.

The feedback mechanisms associated with the increase in surface absorption of solar radiation are often cited as the main
70 contributor to the loss of ice and snow in the Arctic (Serreze and Francis, 2006; Crook et al., 2011). However, Pithan and Mauritsen (2014) proposes that temperature-related processes dominate the Arctic warming. Temperature rise influences cloud formation and precipitation (Herman and Goody, 1976; Curry et al., 1996), which amplify warming in the Arctic region (Taylor et al., 2013), although there are disagreements on the impact of clouds in this respect. For instance Screen and Simmonds (2010) reported that changes in CFC do not strongly contribute to the Arctic Amplification despite their role in “enhanced warming in
75 the lower part of the atmosphere during summer and early autumn”, whereas Francis and Hunter (2006) relate the loss rate of the the perennial sea ice edge shelves to CFC and downwelling LW during spring months.

In addition to the warming from the release of greenhouse gases, the dynamics of air masses and physical properties of clouds may contribute to the tropospheric thermal emission. CFC aside, the optical properties of clouds, such as the optical thickness (COT, τ) and effective radius r_{eff} of droplets/crystals, and liquid/ice water content (LWP/IWP), regulate downwelling
80 LW. Model projections of the century ahead show that Arctic clouds during summer are rather unaffected by sea ice variability but their response to sea ice loss is to become optically thicker, have higher LWP, and more frequent low-level liquid phase within the boundary layer (Morrison et al., 2019). Hence, the changes in τ and thermodynamic phase of clouds enhance or suppress cloud radiative forcing (CRF) at the surface. This behavior has been identified in the continuous surface measurements above the Beaufort and Chuckchi Seas (Shupe and Intrieri, 2004), at Ny-Ålesund, Svalbard (Ebell et al., 2019) and in the data
85 products retrieved from AVHRR (Francis and Hunter, 2006).

From the above review of current knowledge about the changing conditions in the Arctic, we consider that investigations of the R_{λ}^{TOA} and the cloud properties over the past two decades will provide insight into the evolution of the Arctic Amplification. We have prepared a consolidated R_{λ}^{TOA} data set from 1995 to 2018. A key set of satellite sensors record backscattered radiation in the solar portion of the spectrum. Consequently, this study focuses on the months between April and Septem-
90 ber. The Arctic seasons considered are spring, defined for our purposes as April May June (AMJ) and summer, July August September (JAS) (see App. 2.1). The investigation of R_{λ}^{TOA} trends involved analysis of the last twenty years of cloud properties from the observations of AVHRR, retrieved with the latest algorithms (Stengel et al., 2020) (see App. 2.2), which supersede older popular data sets, for which specific errors have been found (Zygmuntowska et al., 2012). We build on the heritage of



the earlier studies describing the Arctic state and extend the trend analyses limited previously to 1982–1999 (Wang and Key,
95 2005b, 2003).

The objectives of this paper are threefold. Firstly, we provide evidence that space-borne measured, spectral R_{λ}^{TOA} is a valuable
indicator of the changing atmospheric composition and surface properties of the Arctic. Secondly, we determine R_{λ}^{TOA} trends
at regional and seasonal scales and reveal unexpected patterns of behavior. Thirdly, we attribute the trends in R_{λ}^{TOA} to changes
in the thermodynamic phase of clouds. Lastly, we quantify the average radiative forcing by clouds and its changes. We relate
100 the latter to the changes in the physical properties of clouds in response to climate change.

2 Data and Methods

The study of the Arctic geophysical domains by remote sensing requires sensors with broad spectral coverage at an adequate
spectral resolution to be able to separate different spectral features of gases, surfaces, liquid water and ice/snow. We define the
spectral reflectance measured at TOA - R_{λ}^{TOA} - as

$$105 \quad R_{\lambda}^{\text{TOA}} = \frac{\pi I_{\lambda}}{\cos(\theta_0) E_{\lambda}^0}, \quad (1)$$

where I_{λ} is the Earthshine, i.e. the upwelling scalar radiance measured at TOA (units of photons \times s⁻¹ cm⁻² nm⁻¹ sr⁻¹), E_{λ}^0
the unpolarized downwelling solar irradiance (photons \times s⁻¹ cm⁻² nm⁻¹) and θ_0 the solar zenith angle in degrees.

Relevant parameters for the R_{λ}^{TOA} analysis are shown in Fig. 1. The left y-axis of Fig. 1-a shows I_{λ} , E_{λ}^0 for a GOME measure-
ment above the Kara Sea, while the right y-axis shows modeled R_{λ}^{TOA} , in satellite perspective, representing the TOA signal for
110 typical Arctic geophysical conditions. Fig. 1-b shows the wavelength dependence, at GOME spectral resolution, of the spectral
reflectance for different surface types. The almost flat Earthshine between 450–800 nm reveals the presence of a cloud deck
or snow surface in the satellite field of view. Ten wavelength bands of spectral width 5–10 nm have been selected satisfying
the following requirements: they are chosen to be similar to those of sensors' channels used in the literature for comparative
purposes. Their coverage from the UV to the NIR shall provide differential sensitivity to individual components of the Arctic
115 atmosphere-surface. Additionally, they should exclude strong absorption by atmospheric trace gases to avoid misinterpretation
of the observed behavior. Two exceptions are the broadband O₃ Chappuis band (525–675 nm) and the narrow O₂ A-band
(centred at 760 nm). The former, even if smoothed at 5–10 nm resolution, has information on the total column of ozone and on
the structure of the upper troposphere and lower stratosphere. Well-mixed gas, such as oxygen, provide valuable diagnostics
about the depth of the atmospheric column, as seen from space. The A-band is used to assess the surface topography in a cloud-
120 free atmosphere (van Diedenhoven et al., 2005) and altitude, geometrical and optical depth of clouds over dark (Rozanov and
Kokhanovsky, 2004; Lelli et al., 2012, 2014) and bright (Schludt et al., 2013) surfaces.

2.1 Reflectance data

To detect changes on daily, monthly, seasonal and decadal scales several measurements per day of adequate spatial resolution
must be made over several decades. The polar-orbiting spectrometer suite comprising GOME, SCIAMACHY and GOME-2

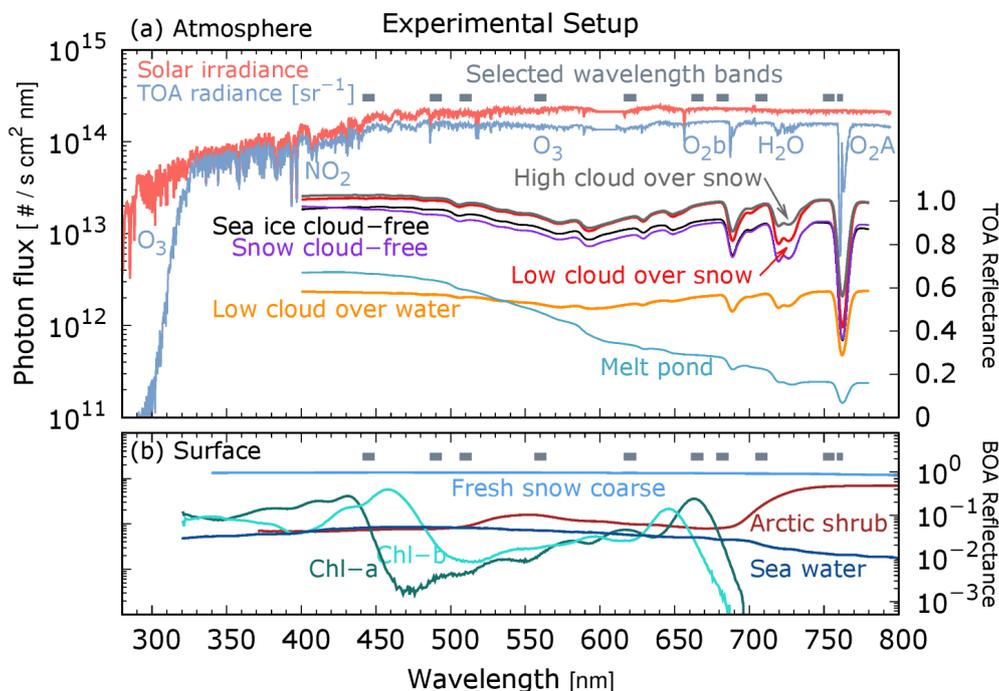


Figure 1. Plots of the solar irradiance, the radiance of a cloud (Earthshine) and reflectances at top (TOA) and bottom (BOA) of the atmosphere as a function of wavelength from 280 nm to 800 nm. The cloud radiance was observed by GOME on May 15, 2001 over Kara Sea (80.53°N, 75.99°E). Modelled R_{λ}^{TOA} (nadir, solar zenith 40°) display a water cloud, placed at 3 km and optically dense 30, above sea water and snow, with a cloud-free sea ice, snow and melt pond spectrum. The lower panel shows the black sky hemispherical reflectance at the ground of relevant Arctic surface components. Chlorophyll absorption is taken from Clementson and Wojtasiewicz (2019) and plotted for a May 2016 concentration of 12 mg m⁻³ observed in the Bering Sea (Frey et al., 2018). Arctic shrub and coarse snow data are taken from the ECOSTRESS and ASTER spectral libraries (Meerdink et al., 2019; Baldrige et al., 2009). Melt pond and sea ice albedos are from Istomina et al. (2013).

125 (Tab. A2 for their specifications) is an optimal choice given the individual length of the time series and their high spectral resolution. Description of GOME can be found in Burrows et al. (1999), while SCIAMACHY and GOME-2 are respectively provided in Burrows et al. (1995) and Munro et al. (2016). The detailed steps to harmonize R_{λ}^{TOA} measured by sensors of different technical specifications are given in the App. A.

130 Sensors measuring solar radiation scattered back to the top of the atmosphere do not measure at night. Measurements in thermal infrared ($\lambda \gtrsim 4 \mu\text{m}$) are required to record the thermal emission from the surface and the atmosphere. In practice, this situation, coupled with different sensor swath widths, limits the use of R_{λ}^{TOA} measurements up to a common north parallel for those months that guarantee adequate sampling. This effect is illustrated plotting the pan-Arctic annual cycle of R_{λ}^{TOA} in Fig. 2. At the three wavelengths 510, 560, and 760 nm, the seasonality shows that summer months have lower R_{λ}^{TOA} and higher otherwise. The darkening of the Arctic can also be seen by comparing the years at the beginning of the record, 1996,

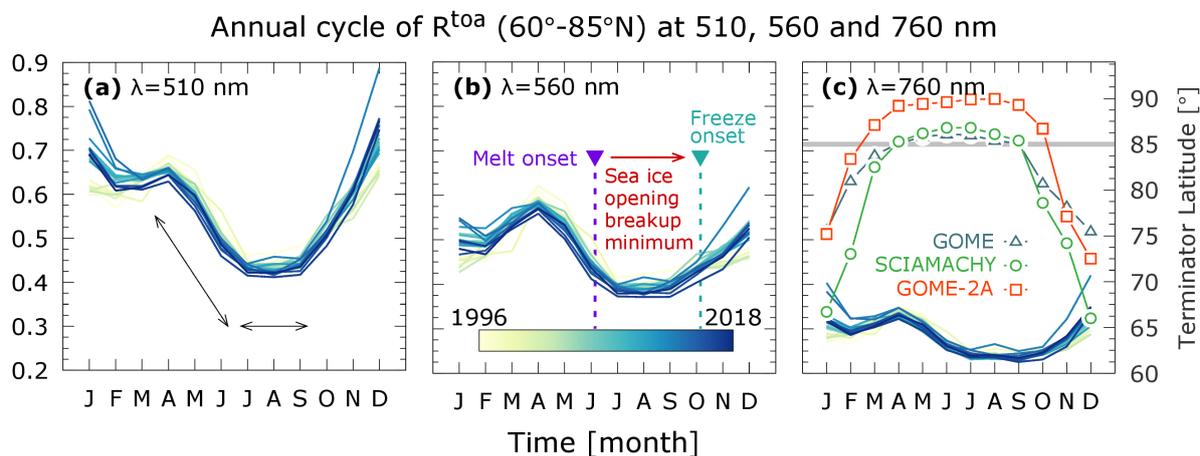


Figure 2. Annual cycle of spectral R^{TOA} at three wavelengths ($\lambda=510, 560, 760$ nm) for the full record from 1996 to 2018. All sets exhibit the demarcation between months of steep (Apr-May-Jun) and flat gradient of R^{TOA} (Jul-Aug-Sep). This shift leads by one month melt onset (6 Jun), followed by sea ice opening, breakup, minimum (16 Jul – Sep inclusive), and freeze onset (4 Oct) as observed with satellite brightness temperatures (Smith et al., 2020). On the rightmost panel the terminator location of the three sensors with the 85°N (grey line) common threshold used for monthly R^{TOA} aggregation.

135 with the most recent ones. However, this behavior occurs between April and September. These are the months when the individual terminator of the three sensors reaches the latitude 85°N , this being the spatial threshold of common coverage. For the other months (October to March inclusive) the individual terminators do not guarantee sufficient sampling (Fig. 2-c) and the R_{λ}^{TOA} curves show that recent years are brighter than those at the beginning of the time series.

From Fig. 2 we identify two distinct behaviors of R^{TOA} . One of steepest decrease, from April to June, and one relatively
140 flat, between July and September. In contrast to the common definition of the climatological seasons, we group April, May, June (AMJ) as Arctic spring and July, August, September (JAS) as Arctic summer. This distinction is explained by the sensors' measurement strategy and by the time-dependent physical processes leading to the transition between high-to-low Arctic reflectance in June to the minimum sea ice extent in September. The changes in surface reflectance between April and May are attributed to snow cover changes and those in June to sea ice changes (Smith et al., 2020). Over water, the timing of such
145 transitions increasingly approaches the summer solstice, which is the day of strongest solar insolation, while it moves further away from it over land (Letterly et al., 2018). It is reasonable to regard this day as a more natural demarcation point between Arctic spring and summer.

2.2 Cloud and flux data

In our study, the R_{λ}^{TOA} data is complemented by a record of cloud properties and fluxes at TOA and BOA. These are inferred
150 from the afternoon orbit of AVHRR sensors onboard the POES missions. The primary reason for choosing these records is the abundance of studies using these data in the Arctic. This has the required coherent radiometric calibration before the



implementation of individual downstream methodology to assess changes across the Arctic. The cloud and flux records, version 3, are presented by Stengel et al. (2020). The application of the cloud algorithm to MODIS measurements, mimicking AVHRR channels, has shown that the retrieval scheme is well aligned with the reference standards of CloudSat and CALIPSO data for CFC, CTH, COT and liquid thermodynamic phase. While agreeing on the sorting of cloud tops between water and ice phases, higher IWP variability is found as compared to that in the reference DARDAR cloud data products (Delanoë and Hogan, 2010). Aggregated IWP histograms do not differ (Stengel et al., 2015). Version 3 has improved version 2 in terms of precision, accuracy and stability (Stengel et al., 2017). Even more relevant to our purpose is the scheme adopted to calculate cloud properties and broadband fluxes. CFC and optical property retrieval uses a neural network trained on CALIOP data to account for the extent of the underlying bright Arctic surface. CTH has been corrected with CALIOP profiles to account for the depth of light penetration inside a cloud. The retrievals of CTH from all infrared thermal channels are influenced by this effect and yield a radiative cloud top height, lower than the physical cloud top. Broadband fluxes are not derived incorporating reanalysis data but the retrieved cloud properties instead. This is important, because the changes in fluxes are coherently related to changes in the cloud properties. Given the standard notation (*all* = all-sky, *clr* = clear-sky, ⁺ = upwelling and ⁻ = downwelling fluxes F), the average long-term relative bias of AVHRR-derived fluxes against CERES ranges from +2.9% for $F_{SW}^{+,all}$ to -2.7% for $F_{LW}^{+,clr}$. Validation with BSRN measurements in the period 2003–2016 shows that the bias (correlation) for $F_{SW}^{+/-}$ in range [-6.16,+1.99] W m⁻² (0.93/0.99) and [-3.02,+7.60] W m⁻² (0.99/0.99) for $F_{LW}^{+/-}$ (Stengel et al., 2020).

Misclassified cloudy scenes especially over dynamically bright surfaces (i.e. marginal and fractional sea ice zones) impact the calculation of broadband fluxes. This has been already noted in first studies comparing ERBE and AVHRR cloud radiative forcing derived with different scene classification schemes (Li and Leighton, 1991). The conversion of directional radiance, measured at TOA, to irradiance requires the knowledge of the angular light redistribution function of the surface and atmospheric components. If this is not accurately assumed, the irradiance and $F_{SW}^{+/-,clr}$ above reflecting surfaces cannot optimally be calculated. Using the same data of our study, it has been found a low sensitivity of trends in cloud radiative forcing to the biases in cloud properties over surfaces of changing brightness (App. d in Philipp et al., 2020, p. 7499). This confirms the suitability of cloud properties retrieved from AVHRR measurements, classified with active sensor data, for Arctic trend studies.

3 Results

The R_{λ}^{TOA} time series, measured by GOME, SCIAMACHY and GOME-2A over the Arctic region (60–85°N), anomalies, trends and significance were harmonized (for more details see App. A and App. B). They are shown for wavelengths 510, 560 and 620 nm in Figs. 3 and 4. The R_{λ}^{TOA} retrieved from the sensors MERIS (on Envisat) and GOME-2B (on MetOp-B) confirm that the correction scheme is successful. A consistent and consolidated data set results from the measurements of the three instruments. Any errors are minimized, when sunlight availability across the Arctic provides full coverage for the sensors' swath at highest latitudes. Seasonality is the dominant feature of Fig. 3. Maximum R_{λ}^{TOA} occurs in early AMJ when the Polar day results in the Arctic being fully illuminated and the ice extent is close to its maximum. Analogously, minimum R_{λ}^{TOA} occurs from August to September when the days are shortening and sea ice coverage is at its minimum. The observed seasonal cycle

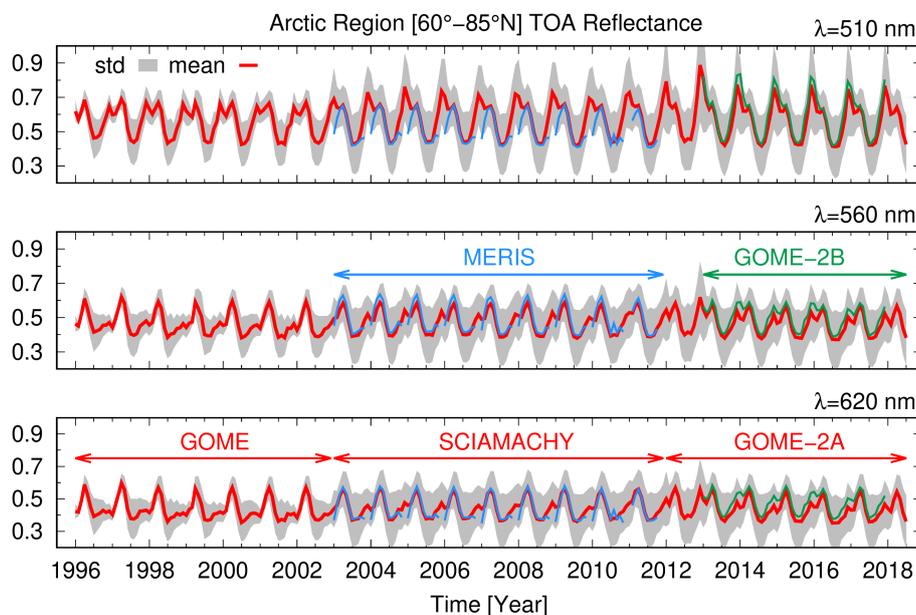


Figure 3. Time series of mean absolute R_{λ}^{TOA} (red lines) and standard deviation (shaded grey) for the three wavelength bands 510, 560 and 620 nm derived from measurements of GOME, SCIAMACHY and GOME-2A over the Arctic Circle. The companion sensors MERIS on board Envisat (blue) and GOME-2B onboard MetOp-B (green) have been superimposed for comparison.

185 of R_{λ}^{TOA} agrees with that observed by models as do the observations of sea ice extent over the Arctic (Holland et al., 2008). This provides evidence to confirm that one dominant parameter in R_{λ}^{TOA} variability is surface reflectance (Sledd and L’Ecuyer, 2019).

A small downward trend of R_{λ}^{TOA} for the three wavelengths in the solar range is seen in the anomalies of Fig. 4. The anomaly of R_{λ}^{TOA} is the difference between the value of R_{λ}^{TOA} and the climatological average value of R_{λ}^{TOA} at time t (see App. B). In a
 190 warming Arctic a significant decrease in reflectance would have been expected due to sea ice loss. For water and ice-covered regions of the Arctic, Pistone et al. (2014) and Morrison et al. (2019) state that no significant relationship between CFC patterns and sea-ice loss is observed during summer but some is identified in autumn months. Such changes are not observable in the pan-Arctic R^{TOA} anomalies. Rather, the reduction in reflectance is small and not attributable to a specific season. As a
 195 consequence, we need to ask whether the loss of reflectance associated with sea ice reduction is compensated by increasing CFC or brighter clouds and which processes lead to the small pan-Arctic R^{TOA} trends.

To answer these questions in the following, we map R_{λ}^{TOA} in the Arctic, gridded at $1^{\circ} \times 1.5^{\circ}$ latitude and longitude. Fig. 5 shows the spatially resolved R_{λ}^{TOA} trends for $\lambda = 510, 560, 620$ nm over the Arctic region for AMJ and JAS. Similarly, Fig. 6 shows trends for the analyzed wavelengths for the 12 Arctic regions, that are defined using the geographical subdivision proposed by Serreze and Barry (2014) and Wang and Key (2005a) (see Fig. B1). Trends for AMJ are shown in green and the
 200 JAS trends for selected spectral bands are shown in blue. The red symbols show the absolute averages of the R_{λ}^{TOA} values at the beginning of the record for the respective seasons.

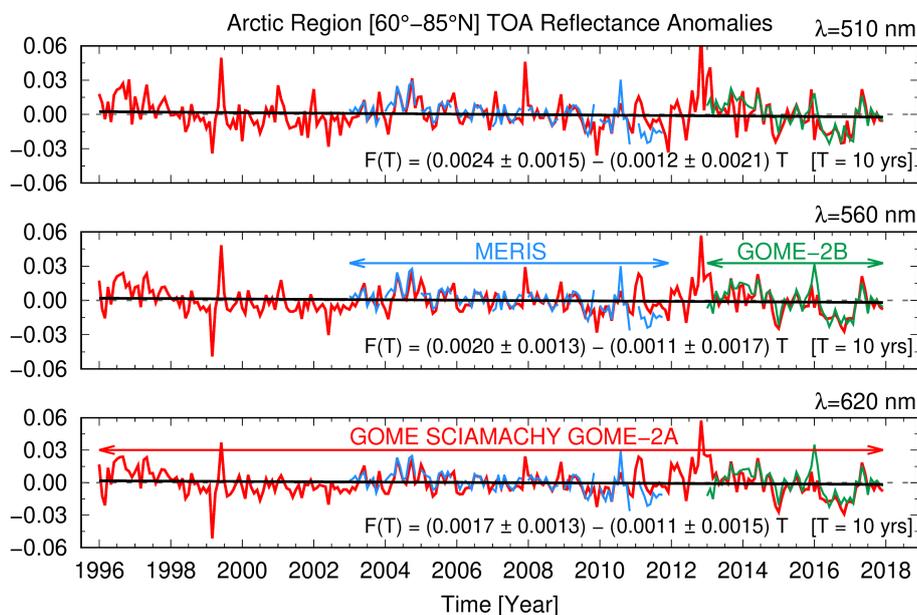


Figure 4. Time series of anomalies of R^{TOA} at $\lambda=510, 560$ and 620 nm derived from the values of Fig. 3. The values are computed with a seasonal cycle on a sensor basis (see Eq. B1). The linear trend $F(T)$ is shown as black line with the bootstrapped intervals at 95% confidence.

There are marked regional differences. Those that are statistically significant (at 95% confidence level) are shown with red crosses. For AMJ a significant negative trend over the Barents Sea is compensated at all four wavelength bands, by a positive R_{λ}^{TOA} trend over Greenland, the Canadian Archipelago and Eastern Arctic Seas. In JAS, the negative trend shifts towards areas of the Kara, Laptev and Chuchki Seas. These are Arctic areas having open ocean and are experiencing significant sea ice loss during the period of study. Statistically insignificant increases in R_{λ}^{TOA} are found over the boreal land masses. However, significant increases in R_{λ}^{TOA} are observed over Greenland and parts of the Arctic Atlantic sector.

Greenland exhibits positive trends of R_{λ}^{TOA} in the visible spectrum in both AMJ and JAS. This is not observed in any other region in the Arctic. The result for the Barents Sea is similarly extraordinary. Here the JAS trends of R_{λ}^{TOA} are strongly negative most likely due to the sea ice loss (Onarheim et al., 2018), but the AMJ result is slightly positive. The values for the most eastern and most western Arctic Seas, i.e. East Siberian Sea, Laptev, Kara, Chuckchi and Beaufort Seas, are often clearly negative for the late summer (JAS) trends, while in AMJ the values show weaker negative or slightly positive trends. Despite its proximity to the Canadian Archipelago, Baffin Bay has changes in R_{λ}^{TOA} trends that would more closely match the Eastern Arctic Seas region. Over the Hudson Bay, the R_{λ}^{TOA} trends show unusual patterns. They are largely positive in JAS and relatively strongly negative in AMJ. During JAS the trends for $R^{\text{TOA}} = 760$ nm are large. We infer that this results from a change in clouds, induced by either increased cloud height (CTH), cloud fractional cover (CFC) or albedo (CA).

Although not of the same magnitude, almost all regions show a reflectance change at $\lambda 760$ nm. Oxygen absorption is modulated primarily by CTH and, to a lesser extent, by CFC and optical properties such as CA and COT. In this context,

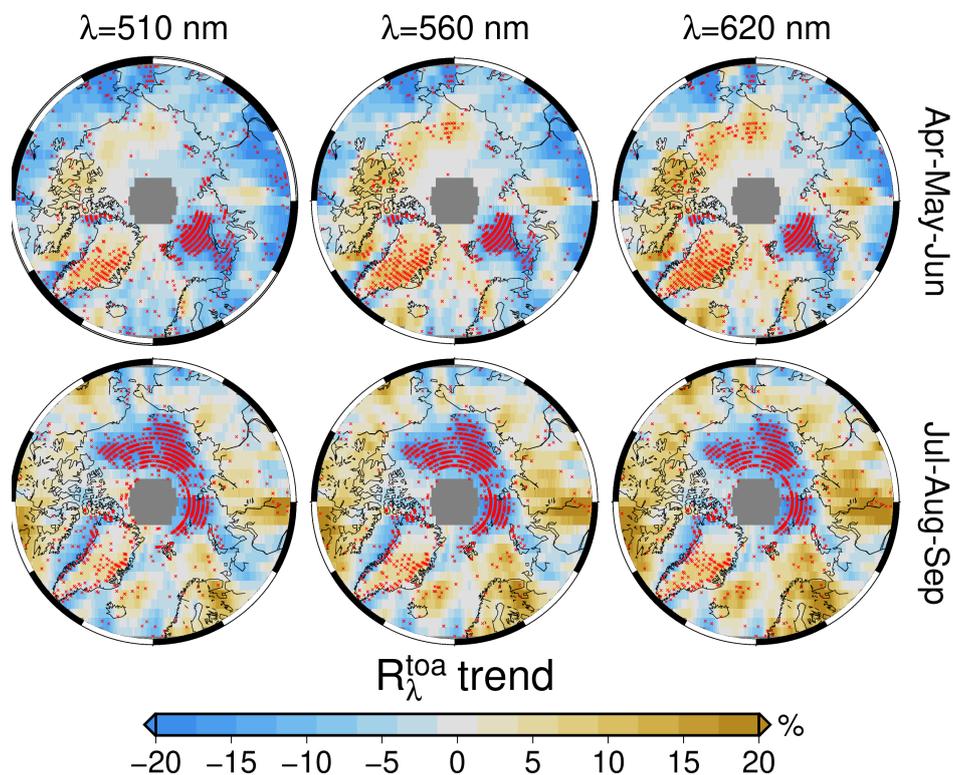


Figure 5. Seasonal R_{λ}^{TOA} trends for 1996–2018 at selected λ for Arctic spring (AMJ, top) and summer (JAS, bottom). The values are relative to the leading season of the record. Stippling in red indicates significant trends at 95% confidence.

a particularly large number of statistically significant large trend values of R_{λ}^{TOA} are observed over Greenland during AMJ, which indicate a clear change in occurrence of clouds or one of their scattering properties. Knowing that R^{TOA} is influenced by scattering and absorption in the atmosphere (Sledd and L'Ecuyer, 2019; Donohoe and Battisti, 2011) and that the atmospheric R^{TOA} can be additionally partitioned into cloud, aerosol and gas contributions, this prompted us to examine changes in those cloud properties which directly influence the spectral R^{TOA} trends.

3.1 Cloud properties

The globally-validated and consolidated cloud record (Stengel et al., 2020) has first been analyzed across the Arctic (60°–85°N). The top plot of Fig. 7 shows time series of CFC and CTH. Both parameters show small, statistically insignificant, trends over the last 20 years. CFC has slightly increased by about 0.001 (+0.14%) decade⁻¹ while cloud tops are lower by ≈ 6 m (–0.14%) decade⁻¹. This finding obviously excludes an explanation being that reflectance loss at visible wavelengths, due to shrinking sea ice extent, is offset by more CFC or that the loss of CFC reveals more bright underlying surfaces. However, the middle plot of Fig. 7 shows that over two decades the COT temporal trend of liquid clouds has opposite sign of that of cold

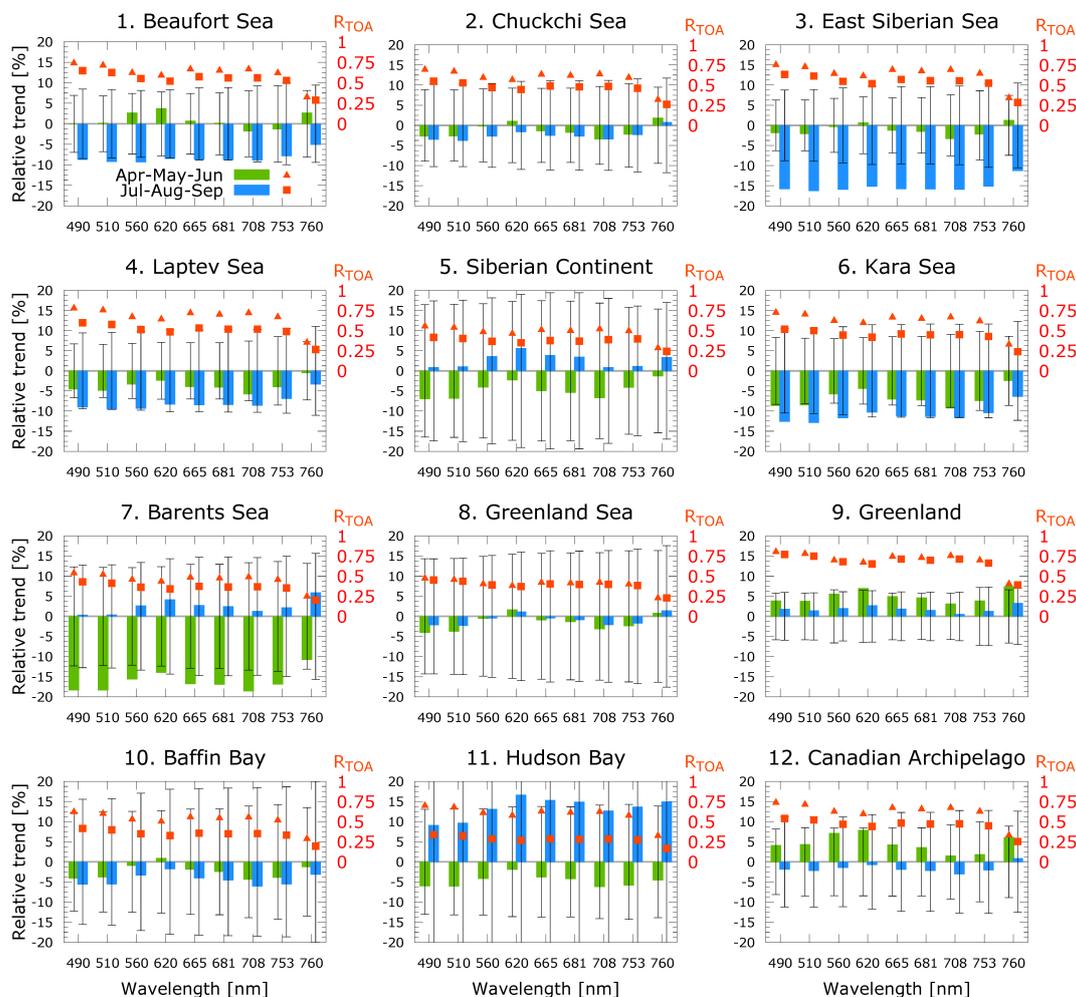


Figure 6. R_{λ}^{TOA} trends for the twelve regions defined in Fig. B1 for spring (AMJ, green bars) and summer (JAS, blue) months. The secondary y-axis display the absolute mean values of reflectance for each Arctic sector. The trend values are relative to the respective lead season and express the total change throughout the record.

ice clouds. τ of liquid warm clouds increases, statistically significantly, by about 0.4 (+2.85 %) decade⁻¹ while the ice-cloud τ decreases by 0.65 (−6.15 %) decade⁻¹ in the same period.

235 Similar to the approach we used for the R_{λ}^{TOA} trends regionally and qualitatively, we map cloud parameters in the bottom panel of Fig. 7, adding also the albedo of clouds at $\lambda = 600$ nm. CFC trends are regionally partitioned and are seen to increase in the range 5–20% where the greatest sea ice losses are observed. This occurs during AMJ and less extensively in JAS. Examples of this behavior are found in the Barents, the Kara and Laptev Seas. On the contrary, large areas of statistically significant decrease in the range 2.5–10% are homogeneously observed across land masses circling the inner polar belt. This includes Greenland and the Atlantic corridor, confirming past results (Hofer et al., 2017). More pronounced trends of the

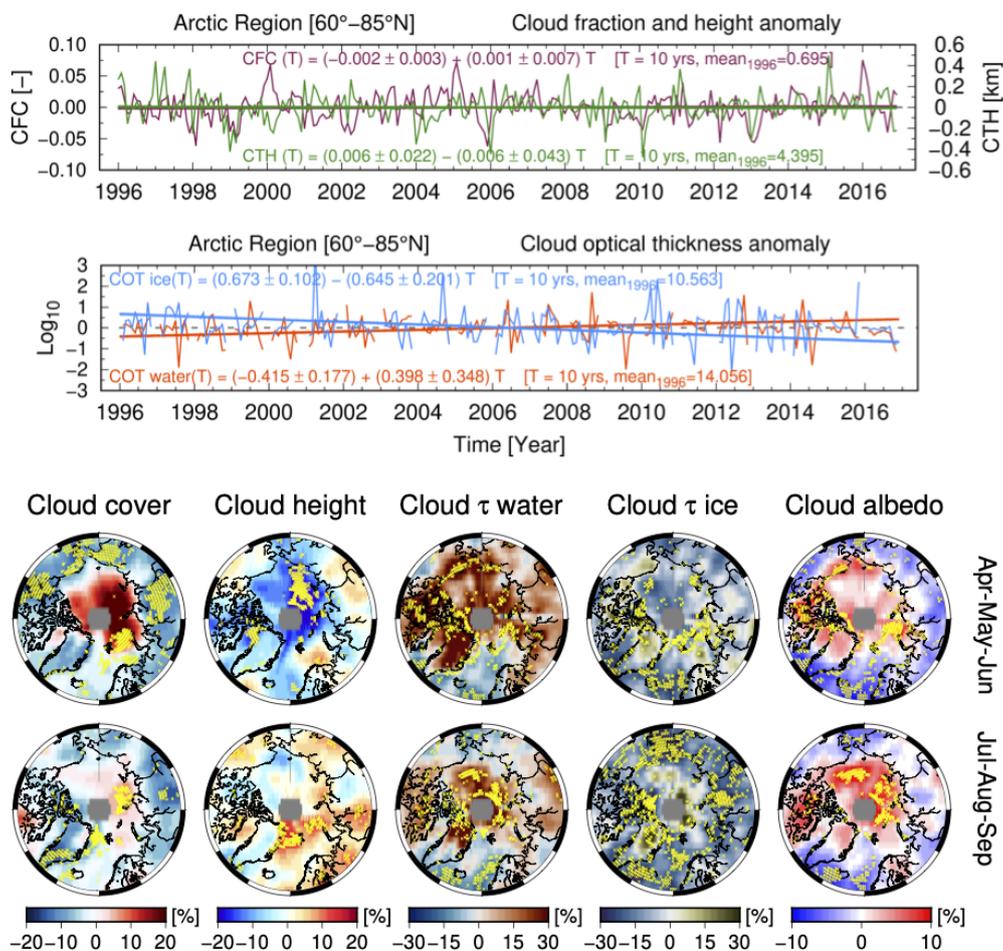


Figure 7. For cloud cover (CFC), height (CTH), optical thickness (COT, τ) of warm liquid and cold ice clouds the left panel shows the pan-Arctic trends, while the right panel shows maps of their seasonal breakdown together with cloud albedo (CA) at $\lambda=600$ nm. The trend values in % are relative to the property value at the start year (1996) in the record. Stippling in yellow indicates statistical significance at 95% confidence.

different cloud parameters, irrespective of their sign, occur in AMJ rather than in JAS. The Hudson Bay is one of the few regions experiencing a seasonal trend reversal. The AMJ period is characterized by less cloudiness (-5%), whereas the JAS period exhibits an increase of the order of almost 10% over the last two decades. The resemblance to the trend reversal of all R_{λ}^{TOA} channels (Fig. 6) indicates that CFC changes primarily modulate R_{λ}^{TOA} over the Hudson Bay. This is inferred from the absence of change of trend sign of those cloud parameters that influence the reflectance in the solar spectrum, such as COT of water and ice clouds (third and fourth column in Fig. 7).

CTH decreases, especially where statistically significant trends are observed, during AMJ across almost all sectors of permanent and marginal sea ice (Beaufort, Chuckchi, East Siberian, Laptev, Kara Seas) and over the Baffin Bay. In the last two

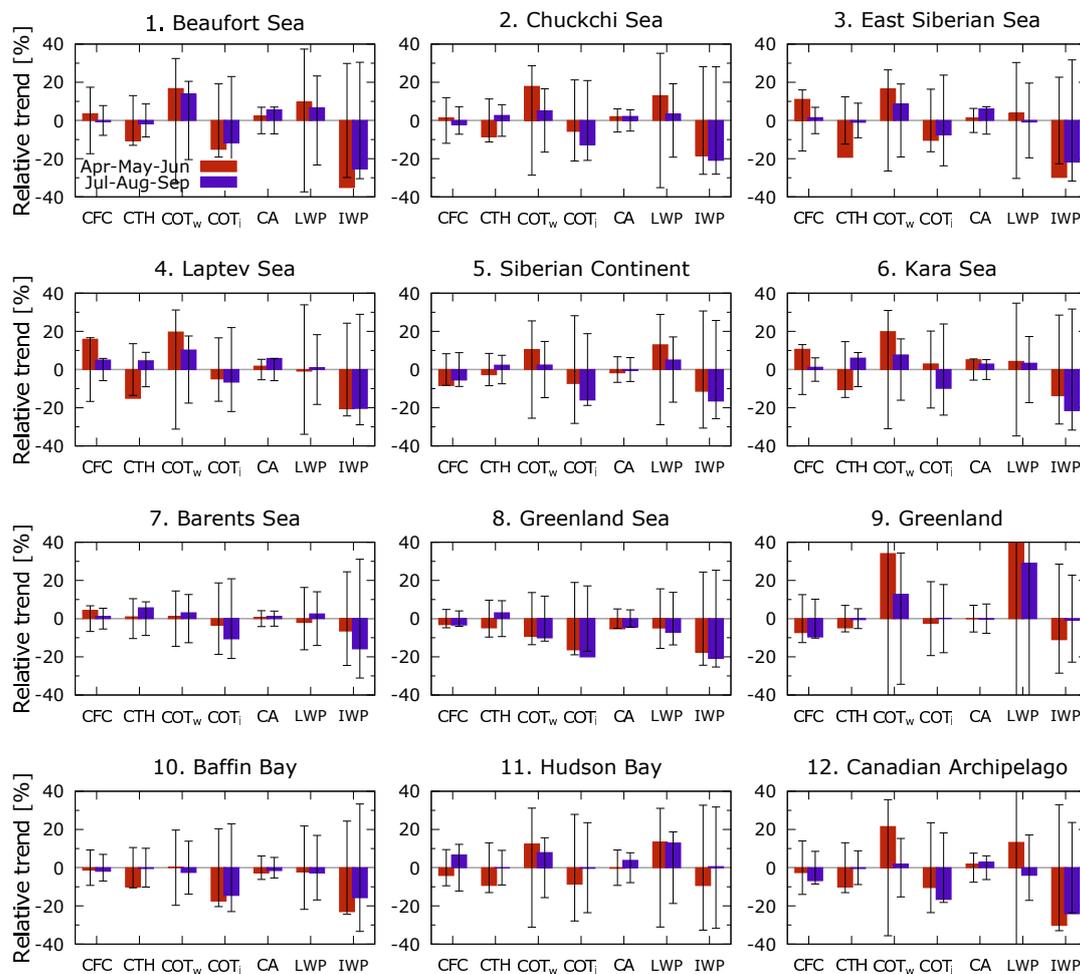


Figure 8. Trends (and $2\text{-}\sigma$ standard deviation) of cloud properties for the twelve sectors defined in Fig. B1 for spring (April - May - June, red bars) and summer (July - August - September, purple) months. The y-axis display the change relative to the leading season in 1996 and express the total change throughout the full record throughout 2016.

decades CTH in these regions has decreased by 10% on average. In JAS, however, CTH increases significantly from the Fram strait, throughout the Barents and Laptev Seas, closer to the Pole, and western Siberia, with a less pronounced positive trend for Greenland (+4%) and a more pronounced one for the Hudson Bay (+8%), albeit non significant. Total COT is split into liquid and solid cloud phases. The geographic distribution of the trends in Fig. 7 provides insight into which areas are responsible for the positive pan-Arctic trend in τ of liquid clouds (τ -water) and for the negative trend for ice clouds (τ -ice). τ -water increases across the whole Arctic in AMJ except over the Atlantic sector and the southern part of the Hudson Bay. The positive trend is maintained over north Greenland, Canadian Archipelago, North Pole and part of the Eurasian continent also during JAS. A positive trend of τ -water is correlated with a trend of opposite sign for τ -ice: this holds for all regions of permanent and



255 marginal sea ice, for the Canadian Archipelago and for the Hudson Bay. Conversely, Greenland, Baffin Bay and the Atlantic
sector show a different behavior: there is a 34% increase in τ -water during AMJ and 22% increase in JAS. Notwithstanding
the increase over localized areas (e.g. north Greenland), mean τ -ice over the Arctic regions remains nearly unchanged in both
seasons. The liquid phase of clouds does not increase across the Fram Strait, whereas the ice phase decreases by roughly 20%
in both AMJ and JAS periods. Finally, the Atlantic sector (the Greenland and the Norwegian Seas) show decreases in the COT
260 for both the liquid and solid cloud phase during AMJ and JAS.

The rightmost polar plots of Fig. 7 show seasonal trends in cloud albedo (CA), for which a marked change of the spatial rather
than temporal scale is observed. To a certain extent, the CA trends are geographically correlated with those of CFC and τ -water.
Individual regions are grouped in a similar manner to the R^{TOA} polar plots: comparable distribution of CA are found over the
most eastern and most western Arctic Seas (Beaufort and Chuckchi, East Siberian, Laptev, and Kara Seas). Positive trends are
265 almost invariably distributed over water masses, the Canadian Archipelago and the northern part of Greenland, irrespective of
the season. In contrast, clouds become darker at lower latitudes, southern Greenland and the Atlantic sector. Over the Siberian
land masses this is not observed, and CA changes in the region, besides being small, are attributed to a competition between
changes in CFC and τ -water. The loss of albedo due to cloud dissipation is compensated by the increment in albedo through
increased τ -water.

270 To facilitate a quantitative seasonal comparison between the Arctic sectors, Fig. 8 shows the trends and the standard error
(i.e. $2\text{-}\sigma$ standard deviation, see App. 2.2) of five cloud properties (CFC, CTH, τ of water and ice phase, CA) together with
trend of liquid (LWP) and ice water path (IWP), from the same cloud record (Stengel et al., 2020). Changes in R_{λ}^{TOA} depend
in the first place on changes in cloudiness and τ (irrespective of the phase), which in turn is a function of LWP, droplet/crystal
effective radius (r_{eff}) and air density ρ (i.e. $\tau = 3/2 \times \text{LWP} / \rho r_{\text{eff}}$). The sign of LWP and IWP trends confirm the τ trends. We
275 infer that τ -water has increased as a result of the positive change of LWP and/or a concurrent systematic pan-Arctic decreasing
trend of r_{eff} (see Fig. C1).

3.2 Cloud radiative forcing

We compute the net radiative forcing, \bar{F}^{cld} or CRF, due only to clouds. The flux data are taken from the cloud record (Stengel
et al., 2020) (see App. 2.2). The difference between F^- and F^+ is the net radiation \bar{F} and $\text{CRF} = \bar{F}^{\text{all}} - \bar{F}^{\text{clr}}$. The multi-year
280 mean and trends of SW, LW and total CRF at the surface are plotted in Fig. 9.

At pan-Arctic scale clouds exert a negative SW radiative forcing of -58.7 and -63.8 W m^{-2} in AMJ and JAS, respectively.
In the same seasons, the LW component amounts to $+46.9$ and $+46.1 \text{ W m}^{-2}$, and the multi-year mean of total CRF is -11.8
and -17.6 W m^{-2} . However, CRF is seasonally and regionally partitioned: clouds' total radiative forcing at the surface is
positive over bright areas as a result of LW offsetting SW. For instance, total CRF over Greenland is $+14.9$ and $+23.5 \text{ W m}^{-2}$,
285 which corresponds to the Arctic sectors over which clouds reflect the least in absolute SW (-19.8 in AMJ and -21.3 W m^{-2}
in JAS) and emit 36.2 in AMJ and 43.3 W m^{-2} LW radiation in JAS. The minimum total CRF is measured over the Baffin Bay,
the Atlantic corridor and Barents Sea in AMJ (-51.1 W m^{-2}) and JAS (-43.4 W m^{-2}). This is explained by the combined
effect of the brighter surface, increasing SW reflectivity and damping upwelling LW, and comparatively low optical COT

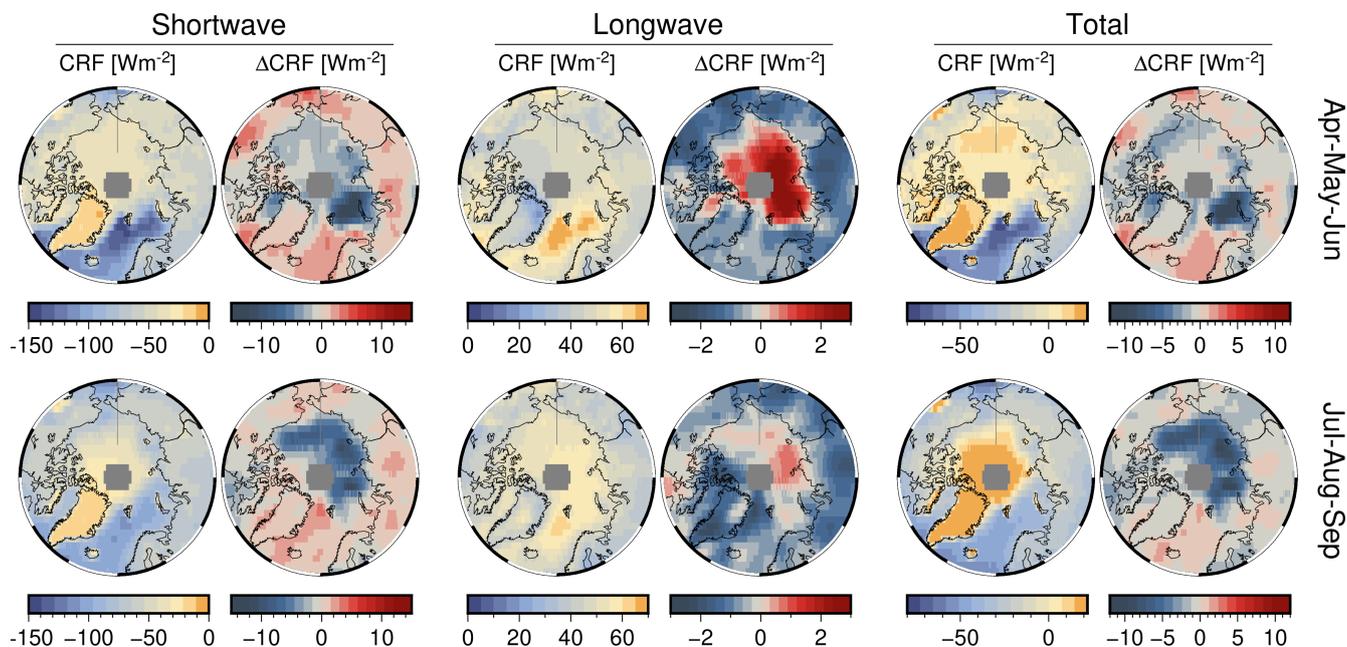


Figure 9. For Arctic spring (AMJ, top) and summer (JAS, bottom), the multiyear mean Cloud Radiative Forcing (CRF) and total change Δ CRF at the surface.

(irrespective of the phase) over Greenland (8.4 ± 7.3 in AMJ and 6.7 ± 3.5 in JAS). For the same seasons, darker surfaces of
 290 the Atlantic corridor and Baffin Bay emit LW more effectively. However, SW offsets LW and total CRF turns negative owing
 to larger τ -water over the Greenland Sea (14.5 ± 3.4 in AMJ and 15.6 ± 3.3 in JAS) or the Baffin Bay (14.6 ± 5.3 in AMJ and
 13.4 ± 3.0 in JAS).

The climatological annual pan-Arctic total CRF (Fig. D1) is positive at BOA ($+9.2 \text{ W m}^{-2}$) with the sole exception of the
 Greenland Sea (-4.2 W m^{-2}). Minimum values are found over Baffin Bay ($+3.3 \text{ W m}^{-2}$) and the Barents Sea ($+5.4 \text{ W m}^{-2}$).
 295 Over Arctic ocean, total CRF amounts to $+7.0 \text{ W m}^{-2}$, which is lower than the $+10 \text{ W m}^{-2}$ reported by Kay and L'Ecuyer
 (2013), while over land masses clouds warm the surface by $+11.0 \text{ W m}^{-2}$. Consequently, the Arctic surface is warmed by
 clouds throughout and our results are qualitatively consistent with the current knowledge (Zygmuntowska et al., 2012; Kay
 and L'Ecuyer, 2013; Intrieri et al., 2002). The maximum cloud warming at BOA occurs over Greenland (AMJ $+14.9 \text{ W m}^{-2}$,
 JAS $+23.5 \text{ W m}^{-2}$) and to a lesser extent above sea ice covered regions in AMJ (East Siberian Sea $+6.9 \text{ W m}^{-2}$, Beaufort
 300 Sea $+5.7 \text{ W m}^{-2}$, Laptev Sea $+2.1 \text{ W m}^{-2}$) and JAS (East Siberian Sea $+0.4 \text{ W m}^{-2}$, Beaufort Sea $+6.5 \text{ W m}^{-2}$). Otherwise,
 the other Arctic regions show a negative total CRF, from -51.1 W m^{-2} over Greenland Sea and -42.2 W m^{-2} Barents Sea
 in AMJ to the -39.2 W m^{-2} over those regions influenced by the climate of low latitudes (Baffin Bay, Greenland and Barents
 Seas) and -30.7 W m^{-2} and -22.4 W m^{-2} over the Hudson Bay and Kara Sea in JAS, respectively.



From the CRF trends of the last two decades (Fig. 9), clouds over the perennial sea ice zone are increasingly cooling TOA
305 (see Fig. D1) and BOA alike, while being neutral to positive over the Atlantic corridor and land masses at low latitudes. In AMJ
months, maximal cooling trends at TOA (BOA) are for Kara and Laptev, up to -2.7 (-2.4) $\text{W m}^{-2} \text{ decade}^{-1}$, and extend along
the Polar Circle up to the northern section of the Baffin Bay through the Chuckchi Sea, albeit dropping in magnitude to -0.9
(-0.8) $\text{W m}^{-2} \text{ decade}^{-1}$. During AMJ, clouds have increasingly cooled the Siberian land masses and the marginal sea ice
310 zones at an average rate of -0.4 $\text{W m}^{-2} \text{ decade}^{-1}$, with the Barents Sea undergoing the strongest CRF drop by -2.5 W m^{-2}
 decade^{-1} . Otherwise, CRF trend at TOA and BOA during JAS varies from slightly positive over land masses, such as Eurasia,
 $+0.1$ ($+0.1$) $\text{W m}^{-2} \text{ decade}^{-1}$, over open waters in the Atlantic sector, the southernmost portion of Baffin Bay, and the Bering
Strait. Cooling trends due to clouds are identified over Greenland for both seasons having a rate of -0.5 $\text{W m}^{-2} \text{ decade}^{-1}$.

4 Discussion and conclusions

In the last two decades, the set of analyzed parameters provides a coherent geophysical picture: the Arctic R_{λ}^{TOA} has declined.
315 However, this decline is less than that expected as a result of the loss of sea ice. We attribute the reason for this decreasing
trend to be a decrease in sea ice, compensated for by wetter Arctic clouds. This results from their increasing water content
and a concurrent simultaneous decreasing ice content. Atmospheric moisture fluxes are increasing as a result of more open
waters and transport (Boisvert and Stroeve, 2015; Rinke et al., 2019). Marked regionality and seasonality of R_{λ}^{TOA} , cloud
properties and CRF across the Arctic is identified in four macro-regions, consistently exhibiting similar behavior: Greenland,
320 the permanent and marginal sea ice areas, the Atlantic sector, and the land masses at lower latitudes.

To some extent, Wang and Key (2005b) anticipate the results of our work. The downward trend in broadband albedo of
 $-1.40\% \text{ decade}^{-1}$ between 1985 – 1999 is confirmed by our negative all-sky R_{λ}^{TOA} trends, implying a sustained sea ice loss
after 2000 and general darkening of the Arctic surface. However, the regional patterns match neither our results nor most recent
knowledge (Hofer et al., 2017). The annual increase of 0.6% in CFC over Canadian Archipelago, Chuckchi Sea and Siberia
325 and, in JAS, over Greenland reported in Wang and Key (2005b) is probably explained by the limited length of the analysed
record. Trends in CFC over Greenland, for instance, level out before 1995 but turn strongly negative afterward, contributing
to a significant loss of the ice shield mass (Hofer et al., 2017). This explains the nonexistent clouds' τ trend in Wang and Key
(2005b), which is in contrast to the significant moistening across most of the Arctic of Figs. 6, 7 and 8.

Greenland has a unique behavior: R_{λ}^{TOA} trends at all wavelengths are positive, irrespective of the season (Fig. 6). The AMJ
330 R_{λ}^{TOA} trends, up to 5%, are even larger than those for JAS. This result is particularly surprising, given the insignificant CFC
trend (Fig. 7,8), thus not contributing to an increase of the overall reflectance. Similar behavior is found in the Hudson Bay and
Canadian Archipelago, which show an increase in reflectance, in contrast to a general darkening of the Arctic. The mechanism
by which these regions increase R_{λ}^{TOA} lies in the link between LWP and CA, through τ -water. In fact, τ -water changes sustain
the correlated R_{λ}^{TOA} changes because of the non-linear relationship of CA to τ -water via LWP. It follows that a R_{λ}^{TOA} loss
335 is overcompensated by wetter clouds in the northern sector and by increased snowfall in the southern part of the Greenland
continent. Cloud LWP has increased by 28-30% over Greenland and by 14-16% over the Hudson Bay and the Canadian



Archipelago, having positive τ -water trends of 30%, 14% and 22%, respectively. Notably, the seasonal behavior of τ -water, increasing over Greenland, is not associated with CFC loss and a positive CRF change in the last 20 years. In contrast, cloud dissipation, increased by anticyclonic activity and concurrent temperature inversion strengths, is responsible for enhanced insolation at the ground and melting (Hofer et al., 2017). JAS R_{560}^{TOA} changes over the Hudson Bay are exceptional. They are correlated with a 9% increase in τ -water and minimal CRF changes. This area shows one of the largest CFC increases during JAS months (Fig. 8), also corroborated by similar significant changes in AMJ and JAS observed in the reanalysis data (Fazel-Rastgar, 2020). The total CRF is -30.7 W m^{-2} while $+2.7 \text{ W m}^{-2}$ during AMJ. CRF trends point to a cloud cooling of the Hudson Bay at a rate of -2.9 (AMJ) and -1.3 (JAS) W m^{-2} over the last two decades.

Cloud forcing at the surface depends on cloud property changes. The behavior is summarized in the seasonal and regional charts of Fig. 10, in which mean value and trend of SW, LW and total CRF are shown as function of τ -water of clouds, LWP and CFC changes. It is evident that the strong relationships in AMJ and JAS between total CRF, COT, and LWP are more important in modulating radiation than CFC. While SW CRF cooling dominates over LW CRF warming, CFC changes modulate mainly the LW portion of cloud radiation in both seasons. Arctic regionality emerges from the clustering of the regions, especially in AMJ and to a lesser extent in JAS. In the last two decades the net energy radiating at the surface due to clouds is decreasing. Clouds cool the surface when the upwelling SW energy dominates the downwelling LW radiation. This is the case when clouds become optically denser and hence more reflective.

Those regions characterised by a darkening surface undergo an increase in SW reflection, leading to an increasing cooling by clouds ($\Delta\text{CRF} < 0$). This takes place over the perenniennial sea ice zone (Beaufort, Laptev and East Siberian Seas), where a CRF decrease at a rate of $-1\text{--}2 \text{ W m}^{-2}$ is associated with greater cloudiness in AMJ and increasing τ -water in JAS. Quantitatively, with values of $\Delta\text{CRF}_{\text{Total}} = -1.4 \text{ W m}^{-2}$ and $\Delta\text{CF} = 3.03 \%$, we obtain the total long-term sensitivity $\Delta\text{CRF}_{\text{Total}}/\Delta\text{CF} = -0.48 \text{ W m}^{-2} \%$ over the Beaufort Sea in AMJ. The sensitivities of the SW and LW parts of CRF amount to -0.56 and $+0.84 \text{ W m}^{-2} \%$. Although averaged over one multi-year season only, our estimation is in line with measurements reported at the same location during the SHEBA campaign (Shupe and Intrieri, 2004). The SHEBA sensitivity of $\partial\text{CRF}_{\text{LW}}/\partial\text{CF} = 0.65 \text{ W m}^{-2} \%$ was seen to offset the SW for most of the year (with $\partial\text{CRF}_{\text{SW}}/\partial\text{CF} \in [0, 1] \text{ W m}^{-2} \%$), thereby warming the surface while cloud cooling took place only in midsummer months. Accordingly, we report a net total (SW+LW) sensitivity of $-0.13 \text{ W m}^{-2} \%$ in JAS, meaning that the SW cooling takes over LW warming during the Arctic JAS in the record. The greenhouse effect from increased CFC in AMJ over these regions is not directly linked to the retreat of sea ice, the onset of which is in late May (Smith et al., 2020). Rather it is attributed to the enhanced convergence of atmospheric water content originating from open Arctic oceans during years with anomalously low sea ice extent (Kapsch et al., 2013). Our results imply that this mechanism is not only evident in the year-to-year variability of exceptional sea ice lows, but is also a long-term component at decadal time scales, during which atmosphere-ocean coupling effects are predominant.

With the sole exception of the East Siberian Sea in JAS where τ -water of clouds grows in spite of a lower content of liquid water ($\Delta r_{\text{eff}} \approx +0.3\%$, see Fig. C1), any positive τ -water trend corresponds to LWP changes for both seasons. Although not surprising, we note that the AMJ changes in CRF do not correlate with either LWP or COT. In the JAS months, however, larger cloud optical densities and LWPs are matched by a decrease in CRF at the surface. This is the combined outcome of

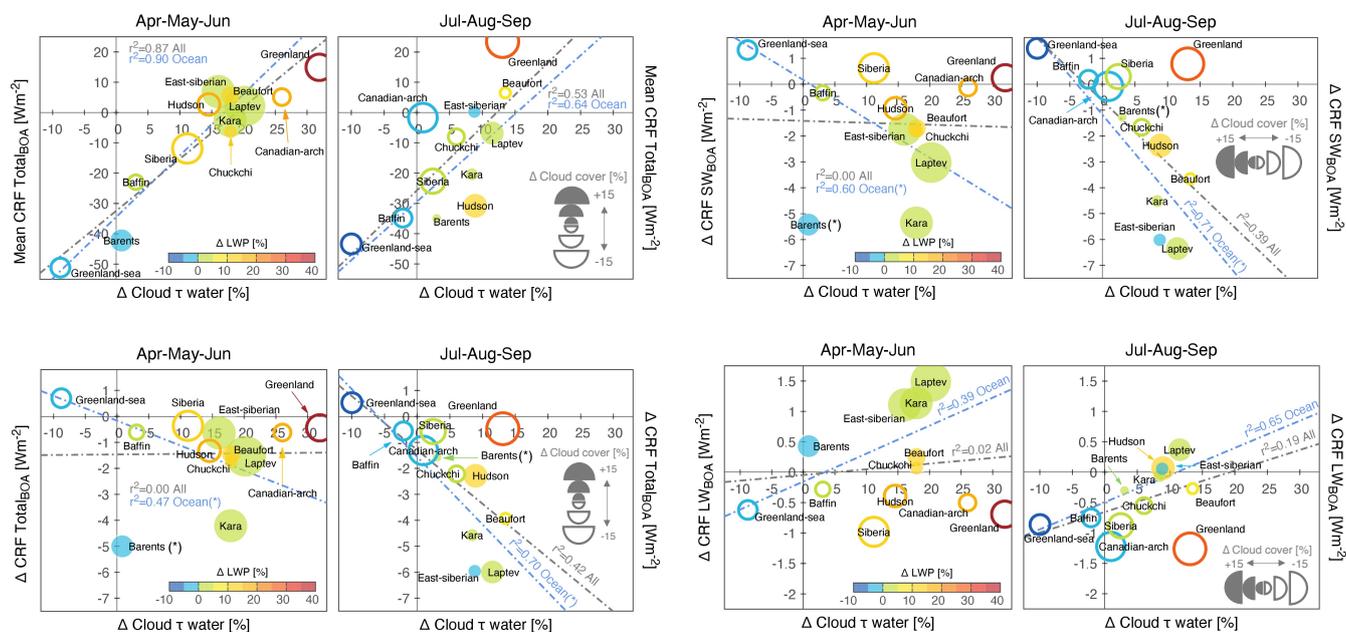


Figure 10. Regional and seasonal mean CRF (top left panel), SW CRF (top right), LW CRF (bottom right) and total CRF (bottom left) trends as function of τ trends for liquid clouds. The concurrent change in LWP is color coded while the increase (decrease) in cloudiness is given by a filled (outlined) circle.

two effects: more insolation in the JAS months results in a more efficient SW scattering by cloud droplets and the darkening of the surface lowers the LWP threshold necessary for the CRF_{SW} to dominate CRF_{LW} . Excluding Barents Sea, the variance of ΔCRF during AMJ is narrower (-4.2 to $+0.9 \text{ W m}^{-2}$) than during JAS (-6 to $+0.4 \text{ W m}^{-2}$). This is evidence for the importance of radiance from the underlying surface, which is larger in AMJ than in JAS.

In addition to loss of clouds, the extended ice melt in Greenland is also known to be reinforced by low altitude liquid water clouds having sufficient opacity to allow the enhancement of downwelling LW flux but also being optically thin enough to allow a significant fraction of the SW flux to pass. This results in the surface being warmed (Bennartz et al., 2013). Such clouds occur within the range of LWP between 10 g m^{-2} and 60 g m^{-2} corresponding to the range of enhanced CRF at BOA. Fig. 8 shows that the increase of cloud τ -water and LWP over Greenland is among the largest throughout the whole Arctic for both seasons. We attribute a reduction in melting to the increased wetting of clouds ($\Delta\text{LWP} \geq 40\%$ at current typical LWP values). Similarly, during AMJ, Greenland and the Canadian Archipelago exhibit a rather small ΔCRF along with a large increase in τ -water and R_{560}^{TOA} . Enhanced surface cooling by clouds is thus less efficient. These regions are predominantly land or land/water mixtures, accordingly modulating the SW-to-LW flux balance.

Advances in observational techniques and process-level research are needed to assess unambiguously the relative roles of temperature T and atmospheric particulate matter in determining cloud thermodynamic changes. In the absence of a systematic, pan-Arctic, aerosol indirect effect due to decreasing trends of ice or cloud condensation nuclei (IN/CCN), higher condensation



rates (i.e. positive LWP trends) of small-sized cloud droplets can only nucleate and grow by a combination of changes in Arctic boundary layer depth within a saturated air volume. Different temperature regimes influence CA changing the $\tau - r_{\text{eff}} - \text{LWP}$ relationship (Tselioudis et al., 1992) and favour droplet growth over condensation rates and vice versa (Lohmann et al., 2000). To this end, the driver of, mostly decreasing, r_{eff} trends (see bottom plot of Fig. C1) remains unclear. r_{eff} size spectrum is modulated by the amount of water vapor and available particulate. While model and satellite data show a general moistening of the Arctic (Rinke et al., 2019; Boisvert and Stroeve, 2015), local on-ground (Graßl and Ritter, 2019) and pan-Arctic satellite (Linlu et al., 2021) evidence of a decrease in total aerosol burden is growing. However, IN/CCN can not be directly inferred from changes of column-integrated extinction of total aerosol load. Assuming a CCN decrease is in contradiction with the r_{eff} reduction via the Twomey effect. Alternatively, we speculate that the change in size spectrum or aerosol type might lead to optimal IN/CCN size and hygroscopicity, although the total aerosol amount has decreased. Satellite-derived single r_{eff} values are only representative of the droplet/crystal population at a level of $\approx 1 - \tau$ from the cloud top (Platnick, 2000). We recommend that the available and relevant spectral observations are exploited (Kokhanovsky and Rozanov, 2012; King and Vaughan, 2012) to generate a pan-Arctic picture of in-cloud $r_{\text{eff}}(z)$ profiles, which would optimally complement surveys based on spaceborne active techniques (Chan and Comiso, 2013; Matus and L'Ecuyer, 2017). $r_{\text{eff}}(z)$ profiles, together with aerosol speciation at high latitudes (Schmale et al., 2021) and cloud bases (Lelli and Vountas, 2018), are essential in two ways. First, they constrain IN/CCN activation, supersaturation and, therefore, cloud particle number concentrations (Zheng et al., 2015; Grosvenor et al., 2018). Second, cloud fields will be more accurately separated according to their phase (liquid, ice and mixed-phase) and layering (low, mid, high-level and multi-layered). We consider our results as upper bounds and more vertical resolution will improve our understanding of the evolution of clouds in the Arctic.

From a modelling standpoint, we can validate past results (Morrison et al., 2019), for which the increase in cloud τ -water and LWP are projected to extend well beyond the middle of the present century. Constraining the cloud microphysics and thermodynamic phase will not only be crucial to project future Greenland melting (Hofer et al., 2019) but also to assess the sign and strengths of total cloud feedbacks (Gettelman and Sherwood, 2016). Given the actual and future Arctic temperatures, ice will be increasingly depleted. Hence, τ -water and LWP will increasingly determine net cloud feedbacks (Bjordal et al., 2020). When the cloud ice phase turns to water a negative feedback is expected due to the offsetting of LW by SW. If climate models do not correctly capture this, i.e they do not incorporate more supercooled liquid than mixed-phase clouds (Lohmann, 2002), unrealistically large amounts of ice result, effectively reversing the sign of the net cloud feedback. We consider that this is one reason, which may explain in part the discrepancy between the atmospheric components (CAM) of the Community Earth System Model 2 (Gettelman et al., 2019, Fig. 2). We note that a CAM5 positive cloud feedback at Arctic latitudes becomes negative in CAM6 as a result of an improved modeling of the cloud phase. Coherently, CAM6 projects a warmer Arctic with increased rainfall rates in JAS at the expenses of snow precipitation (McCrystall et al., 2021), as the outcome of poleward moisture streams and wetter Arctic clouds.

In summary, we have identified expected decreasing and unexpected increasing trends of R_{λ}^{TOA} during the period 1996 to 2018 in the Arctic. We then investigated the possible origins of the unexpected increases of R_{λ}^{TOA} , analyzing cloud optical and physical parameters. We conclude that clouds, through changes in their optical properties explain the increase in R_{λ}^{TOA} and



425 that implies an increasing amount of supercooled liquid cloud droplets. The higher reflectance of clouds results in a more negative radiative forcing at the surface, thereby locally dampening Arctic Amplification. However, cooling by clouds implies the strengthening of the meridional temperature gradient. We expect this will lead to increase the inflow of warmer and moister air masses from the lower latitudes into the Arctic climate. This may then either further decrease Arctic Amplification by generating more supercooled liquid water cloud, or possibly enhance Arctic Amplification by the increase input of warmer air, or some combination of the two. Future model projections of the Arctic climate must take into account these effects to accurately predict the impact of anthropogenic emissions of greenhouse gases and short-lived climate pollutants.



430 Appendix A: Detailed description of reflectance data harmonization

Table A2 shows that overpass time, swath and footprint size differ among the sensors used in this work. These sensors are payloads on satellites which fly in sun synchronous orbits having different equator crossing times. Errors in the R_{λ}^{TOA} in the Arctic arising from the 30-minutes time lag are considered negligible for averaged R_{λ}^{TOA} . Monthly aggregation leads to higher means for finer spatially-resolved instruments than otherwise. Thus, intra-sensor radiometric R_{λ}^{TOA} harmonization is a prerequisite
435 for the creation of calibrated time series and the detection of trends. Different application-dependent approaches have been already employed. Krijger et al. (2007) derives gain correction factors based on the number of cloud-free scenes as function of spatial resolution for maximization of usable trace-gas retrievals. Tilstra et al. (2012) separates the influence of scattering geometry and cloud occurrence to correct SCIAMACHY reflectances for the computation of the aerosol absorbing index at UV wavelengths. Both approaches are not suited for our goal. The former aims at the removal of the influence of clouds, which
440 are a primary component of the Arctic environment. The latter examines instrumental performance in a spectral region that is not of direct interest as a result of potential radiometric degradation of sensors and of higher sensitivity to aerosols, whose radiative effects are comparatively small in the troposphere. Conversely, Hilboll et al. (2013) elaborate a method to explicitly take into account the difference in the ground pixel size and spatial misalignment across sensors. This is achieved by projecting the orbit of one instrument onto that of a second instrument. In our case, we select SCIAMACHY as reference sensor due to
445 its well-calibrated spectral behaviour and because it overlaps with both GOME and GOME-2A. A conservative area-weighted remapping scheme Jones (1999) is employed to derive the factor matrix transforming GOME-2A reflectances as they were measured by SCIAMACHY. Due to the frequent overlaps at high latitudes, only those GOME-2A orbits closest in time to SCIAMACHY are remapped. To extend the time series beyond the loss of Envisat in April 8th, 2012, full SCIAMACHY geolocations, comprising 431 orbits per month, have been used as target tessellation for the rest of the GOME-2A record. The
450 downside of mimicking SCIAMACHY orbits, due to its design of alternating nadir and limb swath states, is the reduction of the GOME-2A sampling rate. This is compensated for in part by the inherently different cross-swath viewing geometries and changes in illumination. GOME projection onto SCIAMACHY has not been implemented. Not only the two sensors overlap for a limited period of six months, but the relatively low sampling rate of GOME would have resulted in suboptimal statistics, even at a monthly scale. Validation has shown that GOME R_{λ}^{TOA} are consistent with those of SCIAMACHY (see Fig. 3 in main
455 text). Remaining intra-sensor inconsistencies that cannot be compensated for, such as changes due to the dynamic radiometric response over dark-to-bright surfaces, will be eventually accounted for by the trend model.

We tested the assumption that bidirectional surface effects do not introduce error in the detection of the temporal trends of R_{λ}^{TOA} by inspecting monthly distributions of the scattering angle throughout the record, separately for each sensor. This is needed because R_{λ}^{TOA} is, by definition, a directional quantity and depends on the scattering geometry. It has been found
460 that the monthly data aggregation of all individual line-of-sights almost fully cover the hemispheric solid angle, incorporating the hot spots of different scene types. The mean value of the scattering angle of 98.48° in 1996 shifts to 98.41° in 2018 for AMJ (-0.08%) and from 97.03° to 96.55° for JAS (-0.51%). These shifts are considered uncritical for this study and do not introduce artefacts in the record.



Appendix B: Trend and significance estimation

465 Trend detection is performed with the same technique for all the variables and parameters in this study. We illustrate the steps with reflectances. Dropping the subscript λ for readability, the R_{λ}^{TOA} , measured by sensor i and aggregated at month t , $Y(t, i)$, are modelled with

$$Y(t, i) = \mu_i C(t, i) + S(t, i) + \omega_i t + \delta U(t, i) + N(t, i). \quad (\text{B1})$$

The $\mu_i C(t, i)$ are the intercept of the regression line, $S(t, i)$ is the seasonal component of the time series, ω_i the desired trend value and $N(t, i)$ the noise residuals embedded in the model after the regression is carried out. The term $\delta U(t, i)$ stands for the product of the level shift δ among the respective sensor records (Hilboll et al., 2013) with the step function $U(t, i)$ needed to concatenate the individual time series at time $T_i(t = 0)$ (Lelli et al., 2014). The seasonality $S(t, i)$ is accounted for by subtracting the average R_{λ}^{TOA} of each month from the respective monthly value. This method is similar to the harmonic expansion in Fourier series, in which the coefficients are derived in a least squares sense. Both methods are equivalent and the choice of one method rather than the other does not introduce significant errors (Mieruch, 2009). The term $\delta U(t, i)$ is embedded by calculating the seasonality separately for each instrument. Its function is to correct possible artefacts due to the different overpass times of the respective spaceborne platforms. While the offsets $\mu_i C(t)$, centred about their mean absolute value at the beginning of the time series, tend to zero upon the anomaly calculation, the last unexplored portion of the data is the noise component $N(t, i)$, in which autocorrelative effects are buried. The R_{λ}^{TOA} time series are persistent in time and the autocorrelation $\rho \rightarrow 0$ for all Arctic regions after one lag. Thus, not all noise components of the record are random and cannot be treated as gaussian. This limits the informative value of any significance test and hinder the detection of trends. Block bootstrap resampling (Efron and Tibshirani, 1993), belonging to the group of nonparametric methods, does not require prior knowledge of the analytical form of the underlying statistics of potentially non-normal data (Mudelsee, 2010). They rest on the block length of the effective independent random sample (Wilks, 1997, Eq. 19). An empirical sample distribution of the trend magnitude ω is then computed scrambling n times the blocks of the original record. The resulting empirical distribution approximates the unknown ω probability density function. This allows to find the $2\text{-}\sigma_{\omega}$ interval needed for a confidence level at 95%. For all locations where the ratio $|\omega/\sigma_{\omega}| > 2$, the trend magnitude ω exceeds natural variability and is termed statistically significant.

Appendix C: Uncertainty propagation in the cloud record and sensitivity

490 The cloud data set is generated using an optimal estimation framework, which allows propagation of random and systematic uncertainties into the pixel-based retrievals. Following Eqs. 2–5 in Stengel et al. (2017), for each location i at time t , we calculate the true variability $\sigma_{\text{true}}(i, t)$ and the uncertainty of the mean $\sigma_{\langle x \rangle}(i, t)$ for the cloud property x from the mean of the squared pixel-based uncertainties $\langle \sigma^2(i, t) \rangle$ and its standard deviation $\sigma_{\text{SD}}(i, t)$. Further, aggregation into monthly averages requires the uncertainty correlation c , or heterogeneity, relating $\sigma_{\text{SD}}(i, t)$ to $\sigma_{\text{true}}(i, t)$. Because c is not known beforehand, setting it to a fixed value is an arbitrary choice that does not account for the spatial and temporal relationship of algorithmic



errors at pixel level throughout wide-scale cloud fields. Hence, we exploit the fact that $\sigma_{SD} \rightarrow \sigma_{true}$ when $c \rightarrow 1$. This holds when the spatial sampling is the highest, thus we scale the number of successful retrievals of the cloud property x to $c \in (0,1]$ and compute the c -dependent $\sigma_{true}(i,t)$ and $\sigma_{\langle x \rangle}(i,t)$. Temporally, both σ_{true} and $\sigma_{\langle x \rangle}$ change as function of c . Seasonal trends of c reveal an overall increase of maximum 3% in AMJ and 1.9% in JAS over the Barents throughout the East Siberian Sea, whereas c over Greenland, Hudson Bay and the Canadian Archipelago exhibits a decrease of 0.6% in both seasons. This translates into a change of $\pm 0.5\%$ and $\pm 0.4\%$ in σ_{true} and $\sigma_{\langle x \rangle}$, respectively. With this approach, the clouds' heterogeneity of the monthly averages is related to retrieval errors predominantly in the spatial but not in the temporal dimension. Limited to an observational analysis of the cloud record, while uncritical for trend assessments only, $\sigma_{\langle x \rangle}$ can be then successively used to label as meaningful those sensitivities of CRF to susceptible cloud property x , whose trend exceeds $\sigma_{\langle x \rangle}$.

505 **Appendix D: Additional description of ozone trends**

R^{TOA} trends at 560 and 620 nm capture the Chappuis ozone absorption band having a broadband maximum centred about 602 nm and two wings stretching between 525 and 675 nm (Gorshelev et al., 2014). Analysing seasonal stratospheric and total column ozone, we are able to determine an effective modulation of R^{TOA} trends by ozone. Ozone data in Fig. A1 are locally derived from GOME, SCIAMACHY and GOME-2A (Coldewey-Egbers et al., 2005) for the total column values (Coldewey-Egbers et al., 2005) and with SCIAMACHY and the OMPS Limb Profiler measurements for the stratospheric column portion (Flittner et al., 2000; von Savigny et al., 2003; Arosio et al., 2019) in the time window 2003 – 2018. The tangent height of 41.3 km is selected due to its highest sensitivity to stratospheric ozone concentrations, which peaks about that altitude. Ozone is produced in the tropics and circulation patterns transport it poleward. It is usually located above the tropopause and its concentrations are higher during winter months and lowest in summer months. Despite its high variability through the year, total ozone trends are generally small in the order of $\pm 1\%$. Focusing on the Arctic, average total ozone is 353 DU and also exhibits a distinct maximum in spring months and a minimum in summer months. The Arctic-wide trend of total ozone is positive by 3.9 DU ($+1.1\%$) decade⁻¹, in line with global values.

Greater significant positive trends, ranging from +4 to +10% decade⁻¹, are found in stratospheric ozone. They are centred above Greenland and stretch out along the 75°N parallel from the Greenland Sea through the Beaufort Sea in spring (AMJ) with a longer tongue over the Siberian Continent in summer (JAS). Contrasting the total with the stratospheric column yields the influence of the tropospheric ozone only. For those locations where the trend in total ozone is absent but positive in the stratosphere, a negative tropospheric trend can be deduced. This mechanism is consistently found above 70°N from the Canadian Archipelago through to the East Siberian Sea, irrespective of the season, together with the sustained positive trend above the Atlantic (the Greenland Sea), the neighbouring Barents Sea and the northern part of mainland Greenland (Gaudel et al., 2020). This reverses in a dipole fashion in JAS, when patterns of positive trends in total ozone are advected southward. In summary, when analysing R_{λ}^{TOA} trends at $\lambda = 620$ nm, and to a lesser extent 560 nm and 665 nm, changes in ozone contribute for those Arctic sectors affected by the meridional dynamics of air masses in which the stratospheric ozone is increasing. The

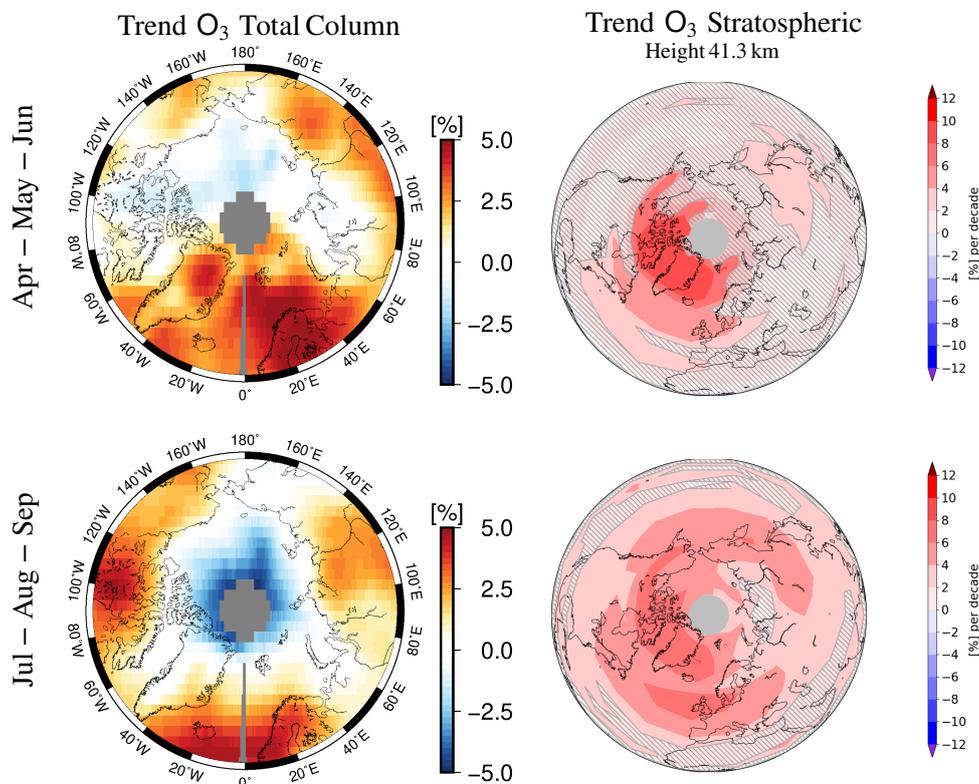
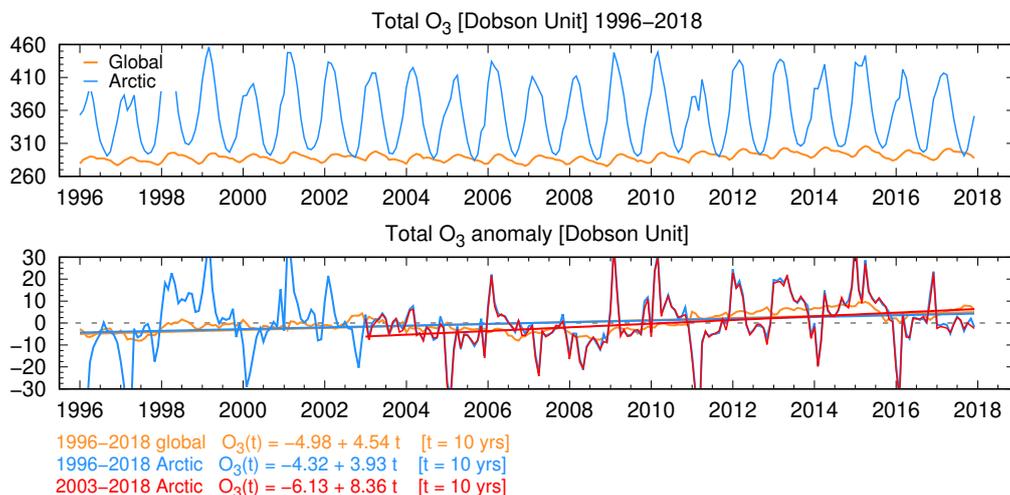


Figure A1. Top: global and Arctic record of total ozone with the respective anomalies and trends. The Arctic time series has been additionally shortened to match the length of the stratospheric ozone column. Bottom: trends (% decade⁻¹) of total (left) and stratospheric (right) ozone between 2003 and 2018 are plotted for spring (AMJ) and summer (JAS) months.



most eastern Arctic sectors (East Siberian, Laptev and Kara Seas) have a smaller contribution from ozone changes than the western sectors. This is consistent with a neutral ozone trend observed over these areas.

530 Finally, we speculate that a surface warming of the Arctic might inflate the tropopause, inducing the production of polar stratospheric clouds as a result of colder temperatures. Lower ozone would absorb less UV and visible radiation, cooling the stratosphere further and potentially accelerating further its depletion. Albeit within natural variability, Turner et al. (2009) held stratospheric ozone depletion responsible for a change in wind flows and patterns across the South Pole, stimulating anti-correlated changes in sea ice extent of the Antarctic continent. This hypothesis could also be tested for the Arctic, using the
535 results from this investigation.



Table A1. List of abbreviations used in the main text.

Acronym	Meaning
AIRS	Atmospheric Infrared Sounder
ASTER	Advanced Spaceborne Thermal Emission and Reflection Radiometer
AVHRR	Advanced Very-High-Resolution Radiometer
BRDF	Bidirectional Reflectance Distribution Function
BSRN	Baseline Surface Radiation Network
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
CALIOP	Cloud-Aerosol Lidar with Orthogonal Polarization
CERES	Clouds and the Earth's Radiant Energy System
DARDAR	raDAR liDAR combined cloud properties retrieval
ENVISAT	Environmental Satellite
ERBE	Earth Radiation Budget Experiment
GOME	Global Ozone Monitoring Experiment
OMI	Ozone Measuring Instrument
MERIS	MEDium Resolution Imaging Spectrometer
MetOp	Meteorological Operational satellite
MODIS	Moderate Resolution Imaging Spectroradiometer
OMPS	Ozone Mapping and Profiler Suite
POES	Polar Operational Environmental Satellite
SBUV	Solar Backscatter Ultraviolet
SCIAMACHY	Scanning Imaging Absorption Spectrometer for Atmospheric Chartography
SeaWiFS	Sea-Viewing Wide Field-of-View Sensor
SHEBA	Surface Heat Budget of the Arctic Ocean
TIROS	Television Infrared Observation Satellite
TOMS	Total Ozone Mapping Spectrometer
TOVS	TIROS Operational Vertical Sounder



Table A2. Specifications of the instruments and data set versions selected for this work. ^aFull coverage until May 2003. ^bPayload switched-off since July 2011. ^cLost contact on April 8, 2012. ^d Nominal end of GOME-2C record. Foreseen extended lifetimes: November 2021 (GOME-2A), 2025 (GOME-2B), 2031 (GOME-2C).

	GOME	SCIAMACHY	GOME-2
Data availability	1996 – 2011 ^{a,b}	2002 – 2012 ^c	2007 – 2023 ^d
Level 1 data processors	5.0	8.01	6.0
Equator crossing (LT)	10:30 AM	10:00 AM	9:30 AM
Global coverage [days]	3	6	1.5
Spectral coverage [nm]	237 – 794	240 – 2400	237 – 794
Spectral resolution [nm]	0.38	0.44	0.48
Pixel size at nadir [km ²]	320 × 40	60 × 30	80 × 40
Swath width [km]	960	1000	1920

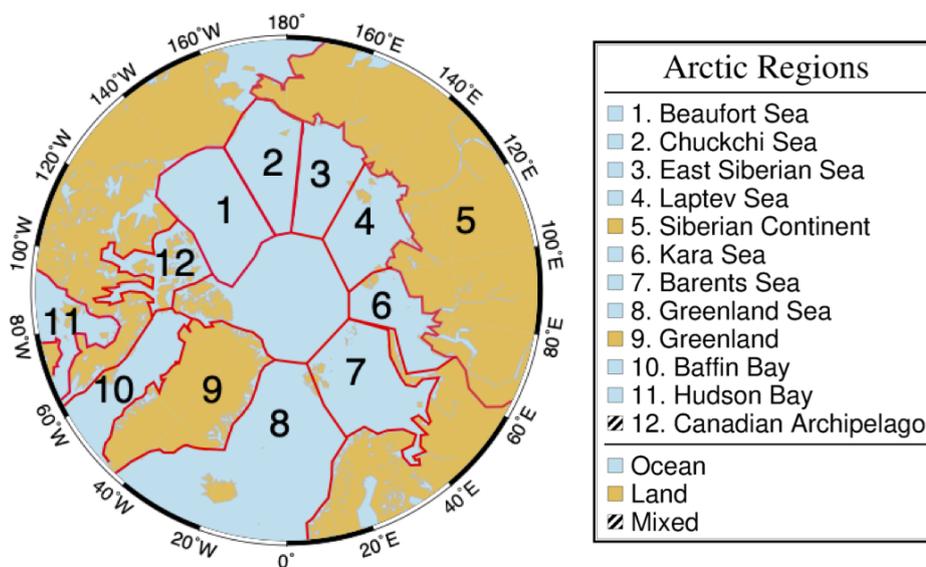


Figure B1. Definition of the Arctic climate zones, identified by distinct geophysical settings, that will be used in this study to derive local trends of R_{λ}^{TOA} , cloud properties and forcing. The geographical subdivision follows that of Serreze and Barry (2014) and Wang and Key (2005a).



Table B1. Multiyear seasonal means (\pm standard deviation) of cloud properties for the full Arctic and 12 regions of Fig. B1.

Region	Cloud cover		Cloud height [km]		τ -water		τ -ice	
	Cloud albedo		r_{eff} [μm]		LWP [g m^{-2}]		IWP [g m^{-2}]	
	AMJ	JAS	AMJ	JAS	AMJ	JAS	AMJ	JAS
Full Arctic	0.70 ± 0.12	0.76 ± 0.10	3.67 ± 0.57	4.14 ± 0.52	13.71 ± 5.75	14.21 ± 3.78	10.34 ± 3.86	12.05 ± 3.80
	0.52 ± 0.05	0.55 ± 0.06	11.87 ± 1.83	12.57 ± 1.43	126.21 ± 64.63	131.56 ± 41.14	148.08 ± 68.71	166.70 ± 73.02
1. Beaufort Sea	0.62 ± 0.19	0.80 ± 0.11	2.82 ± 0.62	3.33 ± 0.48	18.32 ± 8.43	12.71 ± 4.45	12.08 ± 3.67	9.90 ± 3.87
	0.60 ± 0.07	0.58 ± 0.07	10.89 ± 1.83	11.89 ± 1.62	171.35 ± 91.52	111.93 ± 50.43	179.45 ± 81.74	137.50 ± 79.72
2. Chuckchi Sea	0.68 ± 0.14	0.78 ± 0.10	3.30 ± 0.61	3.73 ± 0.50	15.69 ± 7.15	13.91 ± 4.00	10.63 ± 3.63	11.04 ± 3.80
	0.56 ± 0.06	0.58 ± 0.05	11.21 ± 1.91	11.97 ± 1.54	146.07 ± 85.95	123.31 ± 43.60	159.03 ± 73.16	151.06 ± 73.22
3. East Siberian Sea	0.68 ± 0.18	0.82 ± 0.09	2.94 ± 0.64	3.29 ± 0.48	17.43 ± 7.53	13.15 ± 4.13	11.77 ± 3.16	10.89 ± 3.85
	0.58 ± 0.06	0.58 ± 0.07	10.87 ± 1.91	11.96 ± 1.54	157.28 ± 79.00	112.23 ± 41.34	176.44 ± 65.94	153.52 ± 75.82
4. Laptev Sea	0.70 ± 0.18	0.83 ± 0.08	2.99 ± 0.61	3.34 ± 0.46	16.70 ± 7.37	14.77 ± 4.17	12.21 ± 3.34	12.07 ± 4.19
	0.59 ± 0.05	0.61 ± 0.06	10.37 ± 1.93	11.49 ± 1.54	145.73 ± 80.45	122.56 ± 41.38	179.37 ± 70.10	163.62 ± 81.67
5. Siberian Cont.	0.71 ± 0.10	0.74 ± 0.11	4.00 ± 0.55	4.47 ± 0.53	11.67 ± 5.00	15.02 ± 3.69	9.51 ± 4.28	13.02 ± 3.93
	0.47 ± 0.05	0.54 ± 0.06	12.31 ± 1.81	12.90 ± 1.34	106.21 ± 52.39	142.58 ± 40.23	136.00 ± 68.55	183.46 ± 74.40
6. Kara Sea	0.73 ± 0.16	0.82 ± 0.09	3.01 ± 0.62	3.39 ± 0.48	18.22 ± 7.77	16.69 ± 4.35	12.65 ± 3.72	12.80 ± 4.36
	0.59 ± 0.05	0.62 ± 0.05	10.08 ± 1.74	11.35 ± 1.55	151.44 ± 76.56	137.56 ± 38.87	187.25 ± 79.24	167.75 ± 79.36
7. Barents Sea	0.83 ± 0.10	0.84 ± 0.08	2.84 ± 0.47	3.38 ± 0.48	17.25 ± 4.68	17.46 ± 3.77	11.57 ± 3.65	13.31 ± 3.99
	0.59 ± 0.04	0.63 ± 0.04	10.96 ± 1.33	11.81 ± 1.67	141.73 ± 47.12	149.59 ± 36.13	152.60 ± 65.12	170.17 ± 72.77
8. Greenland Sea	0.84 ± 0.07	0.85 ± 0.06	3.18 ± 0.51	3.76 ± 0.59	14.53 ± 3.41	15.65 ± 3.30	10.81 ± 3.16	12.84 ± 3.60
	0.54 ± 0.05	0.58 ± 0.04	12.70 ± 1.31	13.23 ± 1.53	131.02 ± 34.18	147.43 ± 35.89	136.48 ± 51.25	165.13 ± 67.43
9. Greenland	0.51 ± 0.12	0.63 ± 0.11	5.32 ± 0.62	5.42 ± 0.46	8.40 ± 7.33	6.73 ± 3.47	5.97 ± 1.83	5.98 ± 1.83
	0.47 ± 0.05	0.48 ± 0.06	11.23 ± 2.42	11.30 ± 1.55	104.76 ± 134.88	73.46 ± 51.65	99.66 ± 44.58	93.83 ± 36.57
10. Baffin Bay	0.75 ± 0.12	0.78 ± 0.09	3.27 ± 0.60	3.88 ± 0.61	14.65 ± 5.29	13.36 ± 2.98	10.29 ± 3.41	11.64 ± 3.69
	0.52 ± 0.05	0.55 ± 0.05	11.55 ± 1.57	12.94 ± 1.41	129.63 ± 53.41	124.53 ± 32.97	144.34 ± 58.54	157.37 ± 68.47
11. Hudson Bay	0.73 ± 0.12	0.70 ± 0.13	3.33 ± 0.70	4.40 ± 0.64	12.93 ± 5.91	13.04 ± 3.42	9.61 ± 3.82	12.49 ± 4.52
	0.45 ± 0.06	0.51 ± 0.06	11.26 ± 1.84	13.41 ± 1.32	115.37 ± 57.56	123.51 ± 37.05	139.26 ± 62.42	176.42 ± 85.70
12. Canadian Arch.	0.65 ± 0.15	0.78 ± 0.12	3.15 ± 0.69	3.57 ± 0.55	17.24 ± 8.76	13.51 ± 4.15	11.98 ± 4.49	11.44 ± 3.97
	0.57 ± 0.07	0.57 ± 0.06	11.55 ± 1.98	12.52 ± 1.32	174.23 ± 107.76	123.08 ± 46.81	204.37 ± 105.59	162.03 ± 82.11

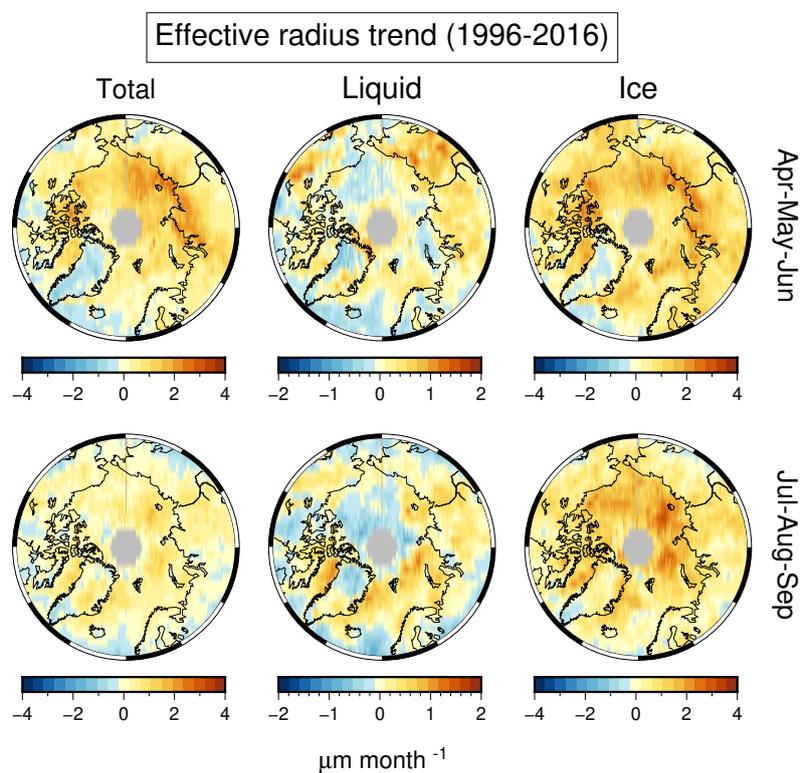


Figure C1. Seasonal trends (top: Apr-May-Jun; bottom: Jul-Aug-Sep) of cloud effective radius (in $\mu\text{m month}^{-1}$) for total cloud (left), liquid only (center) and ice only (right) thermodynamic phase. These values are representative of trends for either liquid droplets or ice crystals located at $\approx 1-\tau$ depth from the cloud top.

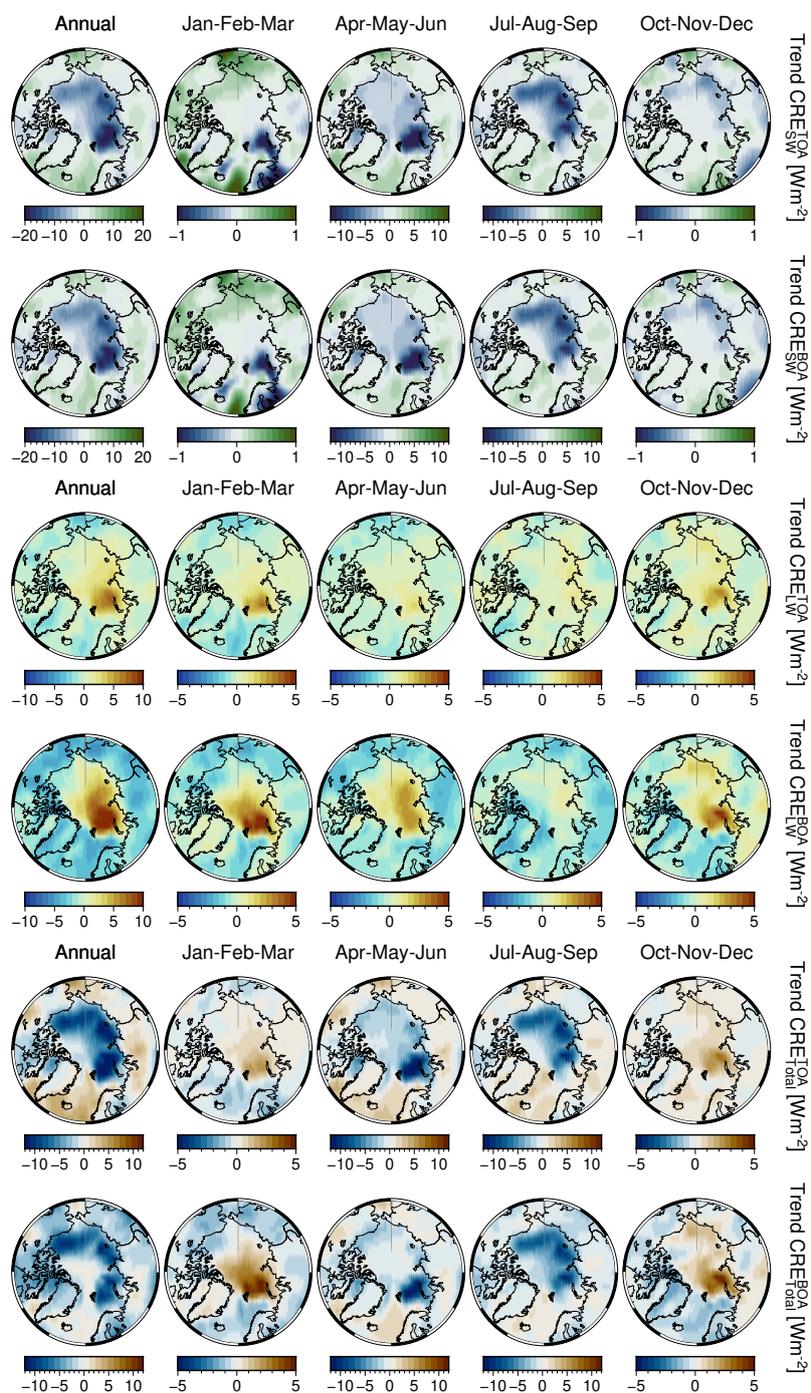


Figure D1. From left to right, annual and seasonal trends of SW (rows 1-2), LW (3-4) and total (5-6) cloud radiative effect (CRE, W m^{-2}) at TOA and BOA.



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540 *Data availability.* Spectral reflectance data are available at <https://doi.pangaea.de/10.1594/PANGAEA.933905>. Cloud and flux data are available at https://doi.org/10.5676/DWD/ESA_Cloud_cci/AVHRR-PM/V003.

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