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Stability dependent increases in liquid water with droplet number in the Arctic

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Abstract. The effects of aerosols on cloud microphysical properties are a large source of uncertainty when assessing anthropogenic climate change. The aerosol-cloud relationship is particularly unclear in high-latitude polar regions due to a limited number of observations. Cloud liquid water path (LWP) is an important control on cloud radiative properties, particularly in the Arctic, where clouds play a central role in the surface energy budget. Therefore, understanding how aerosols may alter cloud LWP is important, especially as aerosol sources such as industry and shipping move further north in a warming Arctic.

Using satellite data, this work investigates the effects of aerosols on liquid Arctic clouds over open ocean by considering the relationship between cloud droplet number concentration (N_d) and LWP, an important component of the aerosol-LWP relationship. The LWP response to N_d varies significantly across the region, with increases in LWP with N_d observed at very high latitudes in multiple satellite datasets, with this positive signal observed most strongly during the summer months. This result is in contrast to the negative response typically seen in global satellite studies and previous work on Arctic clouds showing little LWP response to aerosols.

The lower tropospheric stability (LTS) was found to be the driving force behind the spatial variations in LWP response, strongly influencing the sign and magnitude of the N_d-LWP relationship, with increases in LWP in high stability environments. The influence of humidity varied depending on the stability, with little impact at low LTS but a strong influence at high. The background N_d state does not seem to dominate the LWP response, despite the non-linearities in the relationship. As the LTS is projected to decrease in a future, warmer Arctic, these results show that aerosol increases may produce lower cloud water paths, offsetting their shortwave cooling effect.

1 Introduction

Aerosols can strongly influence the radiative properties of clouds through the modification of cloud microphysical properties. Some aerosols act as cloud condensation nuclei (CCN), and an increase in these aerosols leads to an increase in cloud droplet number concentration (N_d). For a constant cloud liquid water path (LWP), this leads to a decrease in cloud droplet radius (Twomey, 1977). These smaller droplets increase cloud albedo and lead to a shortwave cooling effect. Smaller droplets may also have a larger coalescence rates, and therefore delay the formation of precipitation (Albrecht, 1989). This leads to larger cloud LWP, which also increases cloud albedo. However, an increase in aerosol may also deplete LWP; smaller droplets evaporate more quickly, generating turbulence and accelerating the entrainment of dry air into the cloud (Ackerman et al., 2004; Xue and Feingold, 2006). This promotes further cloud evaporation, which reduces the cooling effect of the cloud.

The size and magnitude of the effects of aerosols on cloud LWP, and therefore net radiative effect, are uncertain. Modelling studies often find increases in LWP with aerosols (Quaas et al., 2008), whereas satellite-based satellite studies typically observe weak or negative responses (e.g., Michibata et al., 2016; Malavelle et al., 2017; Gryspeerdt et al., 2019). Meteorological conditions strongly influence the sign and magnitude of the relationship, with increases in LWP with aerosol loading typically observed in humid conditions (Coopman et al., 2016; Toll et al., 2019).

The relationship between aerosols and Arctic clouds is particularly unclear, in part due to difficulties in obtaining observations (Grosvenor and Wood, 2014). However, as industrialisation moves to higher latitudes, understanding how aerosols change cloud properties will become increasingly important (Schmale et al., 2018). This is particularly essential as low-level liquid-containing clouds play a central role in the Arctic energy budget, in which they often contribute to surface heating through their longwave warming effect (Curry and Ebert, 1992; Shupe and Intrieri, 2004). This contrasts with the rest of the globe, where the shortwave cooling effect dominates (L'Ecuyer et al., 2019). The difference in the Arctic is attributed to two key phenomena; polar night, during which the shortwave cooling effect is non-existent, and the presence of bright surfaces such as snow and ice. Overlying clouds cannot reflect significantly more radiation than these high-albedo surfaces, which again negates their cooling effects. However, Arctic clouds may have a cooling effect in the summer months, when sea ice retreats and there is ample solar radiation (Intrieri et al., 2002). The warming effect of clouds have been linked sea ice loss (Kay and Gettelman, 2009; Huang et al., 2019) and melting of the Greenland ice sheet (Bennartz et al., 2013).

Previous in-situ and satellite studies have shown that Arctic clouds are more sensitive to anthropogenic aerosols than their low latitude counterparts (Garrett et al., 2004; Coopman et al., 2018). The resulting changes in their microphysical properties can substantially change the net radiative effect of the clouds (Lubin and Vogelmann, 2006; Zhao and Garrett, 2015), with the magnitude and sign of the effect also dependent on other factors including season and the albedo of the underlying surface. These modifications in cloud properties may have significant implications for the Arctic, which is undergoing rapid environmental change. The region is warming at an accelerated rate, at least twice the global average (Serreze and Barry, 2011), a phenomenon known as Arctic Amplification. Although primarily driven by an increase in greenhouse gas emissions, clouds play an uncertain role, with models predicting a wide range in the magnitude of their effect (Pithan and Mauritsen, 2014). Aerosol-induced changes to the cloud radiative effects may cause clouds to amplify or counteract this phenomenon (Schmale et al., 2021).

Identifying the role of aerosols on cloud properties is further complicated by the influence of confounding variables. For example, increases in satellite-retrieved aerosol optical depth (AOD) due to aerosol swelling in high humidity conditions may generate spurious correlations between aerosol and cloud properties (e.g., Quaas et al., 2010). Coopman et al. (2016) found that if meteorology is not accounted for, the magnitude of the Arctic clouds response to aerosol is artificially increased by a factor of three. To circumvent these issues, recent studies (Gryspeerdt et al., 2016) have used a mediating variable, such as N_d. N_d is a good choice as its retrieval is not strongly affected by relative humidity and the impact of aerosols on LWP acts through changes to the cloud droplets. By considering N_d, the relationship between LWP and aerosols can be broken down into two

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parts; this can be represented using the sensitivity parameter (Feingold et al., 2001), which quantifies the relative change in LWP for a change in AOD (or N_d):

$$\frac{d\,ln\,LWP}{d\,ln\,AOD} = \frac{d\,ln\,LWP}{d\,ln\,Nd}\frac{d\,ln\,Nd}{d\,ln\,AOD} \tag{1}$$

The use of N_d is particularly helpful in the Arctic; the persistently high cloud fraction (Shupe, 2011; Cesana et al., 2012) and high albedo surfaces means passive sensors (such as MODIS) can only obtain limited valid aerosol retrievals.

Although their high temporal resolution and large spatial coverage overcome the issues faced by in-situ measurements and field campaigns, few previous studies have used satellites to study Arctic aerosol-cloud interactions (Coopman et al., 2018; Zamora et al., 2018; Maahn et al., 2021). This work uses several years of satellite data from multiple instruments to investigate the relationship between LWP and N_d, using reanalysis data for investigate the influence of meteorology. The findings suggest that the lower tropospheric stability (LTS) is a dominant control in the N_d-LWP relationship, which may have significant implications in a warmer, ice-free Arctic.

2 Materials and Methods

Observational data used in this study are obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Aqua satellite for the years 2010 to 2015, inclusive, using the cloud properties from the level 2 collection 6.1 dataset (MYD06_L2; Platnick et al., 2017). Only pixels above 60° latitude were included in this work. The data were regridded from their native 1 km by 1 km resolution to 25 km by 25 km and into the polar stereographic projection. The analysis is performed at an orbital level to avoid temporal averaging of the data. The data were filtered to include only single layer liquid clouds using the 'Cloud_Phase_Infrared', 'Cloud_Phase_Optical_Properties' and the 'Cloud_Multi_Layer_Flag'. Liquid-topped mixed-phase clouds are common in the Arctic, and the MODIS cloud phase algorithm may incorrectly classify these clouds as purely liquid clouds. As such, only pixels with cloud top temperatures above 268 K were included in this study, as in situ measurements show these clouds have a liquid water fraction of upwards of 95% (de Boer et al., 2009).

The cloud liquid water path was estimated according to Equation 2:

$$LWP = \frac{5}{9}\rho_w \tau_c r_e \tag{2}$$

in which ρ is the density of water and τ_c and r_e are the cloud optical thickness and the cloud droplet effective radius, both acquired from MODIS. Equation 2 assumes adiabatic conditions, such as the cloud interacting with the environment through precipitation or entrainment (Brenguier et al., 2000; Wood and Hartmann, 2006).

For comparison with the MODIS data, LWP data was also obtained from version 2 of the Advanced Microwave Scanning Radiometer for EOS (AMSR-E) ocean product, which is also aboard Aqua (Wentz and Meissner, 2004). Data from 2010 and 2011 were included in the analysis. The data were regridded to the same 25 km by 25 km polar stereographic grid as the MODIS data. The in-cloud LWP is calculated by dividing the AMSR-E LWP by the MODIS liquid cloud fraction.

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The cloud droplet number concentration (N_d) is estimated from MODIS data using Equation 3:

$$N_d = \gamma \tau_c^{\frac{1}{2}} r_e^{-\frac{2}{5}} \tag{3}$$

in which the coefficient γ encapsulates atmospheric conditions and is approximately $1.37\ 10^{-5}\ \mathrm{m}^{-1}$ (Quaas et al., 2006). As with LWP, Equation 3 relies on the adiabatic assumption. Marine stratiform clouds are generally found to be close to adiabatic (Zuidema et al., 2005).

Conditions in the Arctic, such as high solar zenith angles and the presence of sea ice, can introduce challenges to obtaining reliable satellite retrievals (Grosvenor et al., 2018). However, carefully filtering the L2 pixels to remove cases in which the data are known to be highly uncertain can prevent the introduction of biases into the results. As such, to limit these uncertainties, the pixels were filtered only to include those with a 5 km cloud fraction of above 0.9 to limit uncertainties associated with retrievals at the cloud edge. Pixels with a heterogeneity index ('Cloud_Mask_SPI') above 30 were removed, as inhomogeneous clouds are known to introduce retrieval biases (Zhang and Platnick, 2011). The solar zenith angle and the sensor viewing angle were limited to 65° and 50° respectively, as high angles are associated with retrieval biases (Grosvenor and Wood, 2014).

Due to uncertainties associated with retrievals of cloud properties over snow and ice covered surfaces by passive sensors, only ocean pixels were considered. The sea ice pixels were removed using daily sea ice cover data from Nimbus-7 SMMR and DMSP SSM/I-SSMIS Passive Microwave Data, Version 1 dataset (Cavalieri et al., 1996), also gridded at a 25 km by 25 km resolution. Open ocean pixels adjacent to sea ice-containing pixels were also removed from this analysis to minimise the impacts of undetected sea ice.

The N_d retrievals were further limited to include only pixels with an r_e greater than 4 μm and a τ_c greater than 4. This is due to uncertainties associated with retrievals of smaller values (Sourdeval et al., 2016). This stringent filtering is not applied to the LWP retrievals as N_d is more sensitive to inaccuracies in these values (following Gryspeerdt et al., 2019).

Meteorological reanalysis data was obtained from the ERA5 dataset (Hersbach et al., 2020), produced by the European Centre for Medium-Range Weather Forecasts. The data at the time step which is closest to that of the time of the satellite overpass is considered to be temporally coincident for this study. The data were gridded onto the same 25 km by 25 km grid as the MODIS data. The effects of free tropospheric moisture and lower tropospheric stability (LTS) were studied as previous works have shown these variables have a strong influence the N_d-LWP relationship (Chen et al., 2014; Michibata et al., 2016; Coopman et al., 2016). The specific humidity at 750 hPa (q_{750}) was chosen as a measure of the humidity of the free troposphere. The LTS, which is a measure of the static stability of the atmosphere, was calculated as the potential temperature difference between 700 hPa and 1000 hPa (Klein and Hartmann, 1993).

Additionally, the marine cold air outbreak index (MCAO; Kolstad and Bracegirdle, 2008) is important to cloud formation and behaviour at high latitudes (McCoy et al., 2017). Much like LTS, it is a metric of the stability of the boundary layer, although it is calculated as the difference between the potential temperature at 800 hPa and the sea surface temperature (Fletcher et al., 2016), with positive values indicating higher instability. This metric is particularly suitable for the Arctic as it highlights the

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difference in temperature of the relatively warm ocean with the cool overlying air masses. The ocean-air temperature gradient is an important driver of boundary layer instability in the Arctic (Kay and Gettelman, 2009).

3 Results

3.1 The regional and seasonal N_d-LWP relationship

Figure 1 (a) shows the annual mean linear sensitivity of the MODIS LWP to the N_d. There is a clear negative-to-positive gradient in the sensitivity, with increases in LWP with N_d typically occurring at higher latitudes. The seasonal cycle of the MODIS sensitivity is shown in Figures 1 (c), (d) and (e). Although spring and autumn have regions of positive sensitivity, the summer months most strongly contribute to the signal observed when considering the all-season response, as they have the most data. Figure 1 (b) shows the linear sensitivity between MODIS N_d and AMSR-E LWP for all seasons. The positive relationship and spatial pattern is similar to the MODIS data, with the AMSR-E LWP show a stronger positive response to aerosols. This may be due to a potential negative bias in the MODIS data due to retrieval errors (Gryspeerdt et al., 2019). Due to the absence of incoming solar radiation during polar nights, there is a lack of data for the winter season.

The decrease in LWP with N_d at lower latitudes has previously been observed in liquid-phase clouds over subtropical oceans (e.g. Michibata et al., 2016; Gryspeerdt et al., 2019) and is consistent with the mechanism of aerosol-enhanced entrainment mixing (Ackerman et al., 2004). Positive sensitivities have previously been observed in high relative humidity conditions (Chen et al., 2014; Toll et al., 2017). Increases in LWP also occur in very clean conditions due to precipitation suppression in this low N_d regime (Gryspeerdt et al., 2019).

There are several possible explanations for the spatial heterogeneity in the LWP response. One potential cause could be due to air masses moving off of the ice edge; cold air moving over the relatively warm open-ocean regions has previously been associated with Arctic cloud formation (Pithan et al., 2018). However, the lack of a strong positive response around the ice edge during the spring months (Figure 1 (c)) suggests that the exposure of these air masses to new aerosol and moisture sources as they transition from the ice pack suggests this is not significant to the N_d-LWP relationship. The remaining potential drivers include the cloud background state and meteorological conditions, which are investigated further in this work.

3.2 The role of meteorology and cloud background state

To investigate the drivers of the different LWP responses, Figure 2 shows the distributions of cloud microphysical properties and meteorological variables for the positive and negative sensitivity regions. There is no evidence for a statistically significant difference between the distributions of the LWP, specific humidity and N_d for two regions (Mann Whintey U test, $p > 0.05$) . However, the distributions for the LTS, the surface temperature and the MCAO index are significantly different. In particular, in the region of positive sensitivity, the LTS tends towards higher values, whereas the surface temperature and the MCAO index are both lower. This influence of stability on the LWP sensitivity is consistent with previous studies (Chen et al., 2014); higher

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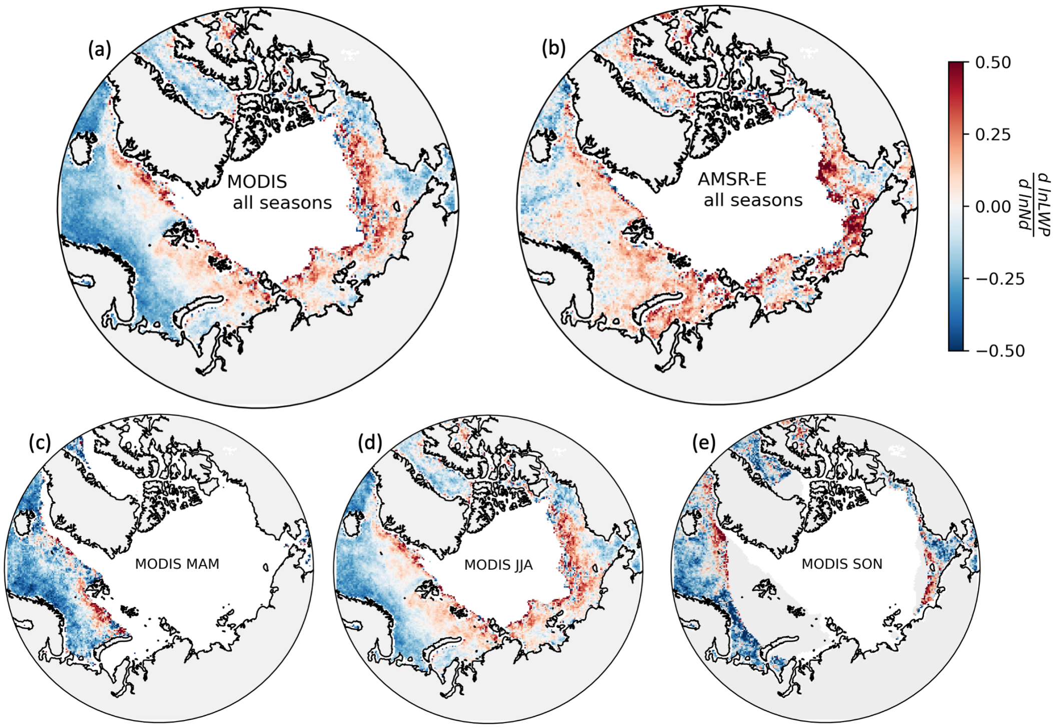

Figure 1. The sensitivity of the cloud liquid water path to N_d for (a) MODIS all seasons, (b) AMSR-E June, July and August, (c) MODIS March, April and May, (d) MODIS June, July and August and (e) MODIS September, October and November. White indicates data omitted due to the presence of sea ice while grey shows either missing data due to polar night or the presence of land masses.

stability conditions inhibit mixing between the cloud layer with the dry above-cloud layer, and prevents the depletion of LWP due to the evaporation-entrainment mechanism.

The r^2 values of the correlation between the sensitivity is higher for the mean LTS (0.39) than for the MCAO index (0.26) and surface temperature (0.32). This indicates that the LTS explains a greater fraction of the variance in the sensitivity than the other meteorological variables. Due to its better performance as an explanatory variable, LTS is used in the remainder of this study as a proxy for the importance of stability and surface forcing on cloud formation.

This association between positive sensitivities and LTS explains why the strongest positive signal was observed during the summer months. The Arctic boundary layer experiences high stability during the summer months, as the ocean temperature is limited by melting sea ice, but the overlying atmosphere can warm (Persson, 2012; Kay and Gettelman, 2009). This creates an ocean-air temperature gradient and high LTS conditions. During the autumn, the ocean's thermal inertia means it maintains relatively warm temperatures relative to the air, thereby decreasing atmospheric stability.

From Figure 2, it appears that the specific humidity does not strongly drive the LWP response. However, previous work has shown that specific humidity often strongly influences the N_d-LWP relationship, with more weakly negative responses under

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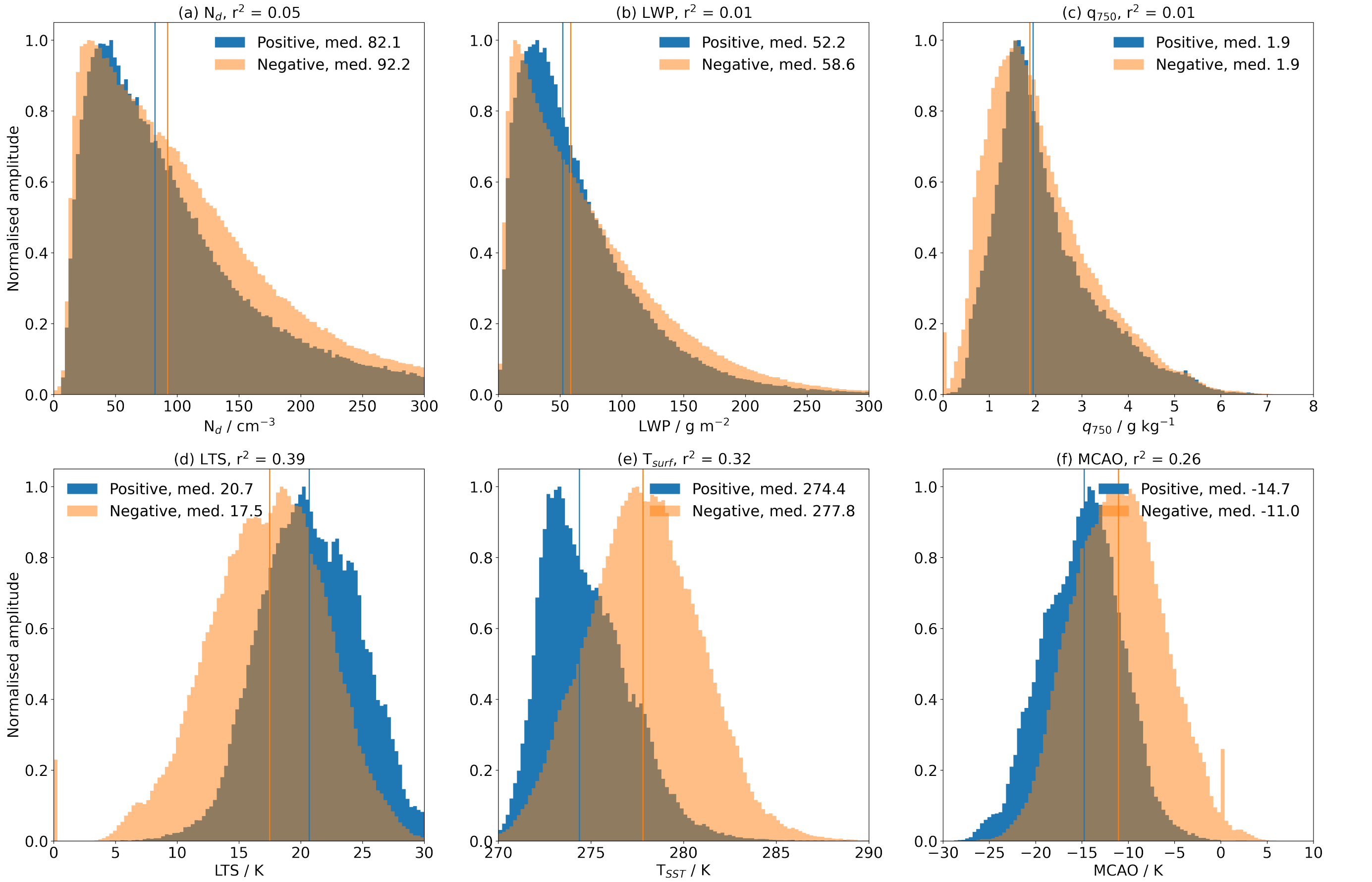

Figure 2. Normalised histograms showing the distributions of (a) N_d, (b) LWP, (c) LTS, (d) q, (e) T_{surf} and (f) MCAO for the regions of positive and negative sensitivity shown in Figure 1 (a). The blue and orange vertical lines show the medians for the positive and negative regions, respectively.

higher humidity conditions (Chen et al., 2014; Toll et al., 2019) due to a suppressed evaporation-entrainment mechanism. To investigate this further, the data were partitioned into bins of specific humidity and LTS (Figure 3). The response to changes in specific humidity is weak, with strongly negative sensitivities evident under both humid and dry conditions. The response to variations in LTS is greater, with a strong negative response at low LTS turning into a positive sensitivity in higher stability conditions. The strong dependence on stability supports the hypothesis that the LTS is the predominant driving force behind the differences between the regions of positive and negative sensitivity (Figure 2).

Although the overall response to humidity is weak, Figure 3 shows that its role is dependent upon the LTS conditions. The influence of humidity is small at low LTS, with a strong negative response across the humidity range. However, it becomes important at high LTS, where the response changes from negative to positive as humidity increases. These results are similar to

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previous work on Arctic clouds; in high stability environments, (Coopman et al., 2016) found an increasingly positive response with q_{750}. Additionally, the sensitivity to aerosol increased with LTS when q_{750} was constrained between 2.0 and 4.0 g kg^{-1}.

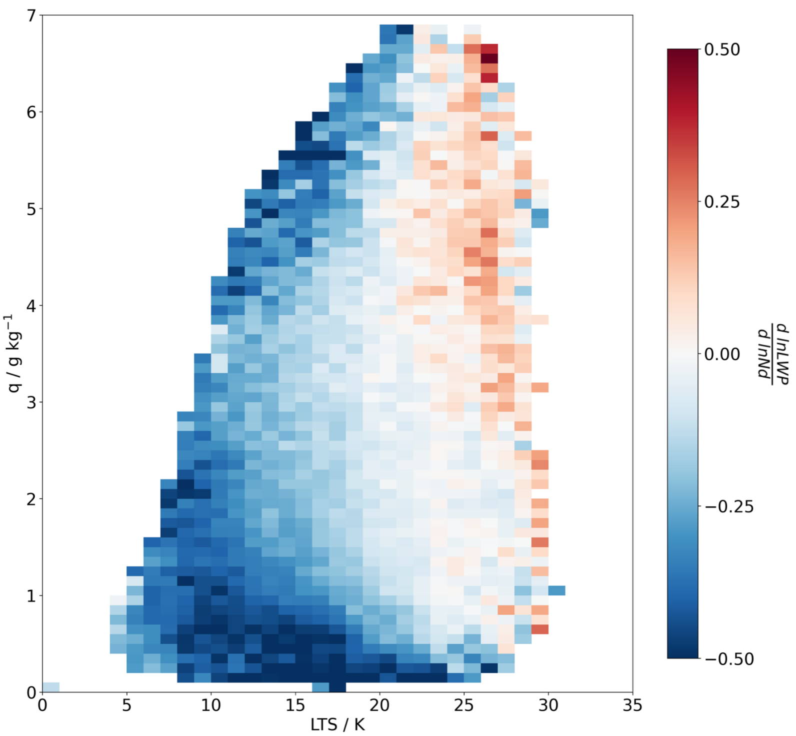

Figure 3. The linear N_d-LWP sensitivity plotted as a function of LTS and cloud-top humidity (q_{750}).

The results presented so far have assumed a linear sensitivity of LWP to N_d; however, assuming linearity means important characteristics of the relationship are not considered. For example, the response of LWP can be non-linear, with a dependence on the initial cloud state (Gryspeerdt et al., 2019). Additionally, use of the linear sensitivity parameter does not consider the absolute values of the LWP and how these change in different meteorological regimes. To investigate these characteristics, joint probability histograms were generated by creating a 2D histogram of LWP and N_d and then normalising each column by the total N_d, such that each pixel in that column represented P(LWP|N_d), or the probability of observing a particular LWP given a particular N_d. These diagrams also allow for exploration of where along the N_d spectrum the meteorological conditions considered in Figure 3 become important to the N_d-LWP relationship.

Figure 4 shows the N_d-LWP joint probability histogram for four different environmental regimes, partitioned into high and low LTS and specific humidity bins. The differences between the normalised histograms are shown at the end of each row and column, and the N_d distributions for the regimes are shown beneath each histogram. The black lines are the mean LWP for each N_d bin, with the shading showing the 95% confidence interval. The blue lines on the difference plots shows clouds with droplet effective radius of 15 μm; clouds to the left of this line are assumed to be precipitating.

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The top row in Figure (4) shows that the LWP increases with N_d under high LTS conditions, as expected from the previous results considering the linear sensitivity. Stronger increases occur in cleaner (low N_d) background states, as has been observed in previous work (Gryspeerdt et al., 2019); this is consistent with precipitation suppression. However, the N_d distributions below the histograms show that these very low N_d conditions are relatively rare in the Arctic and therefore have little impact on the linear sensitivity. There is little difference in the high and low q_{750} environments in these low N_d conditions, suggesting that the precipitation suppression mechanism is not strongly reliant on specific humidity. However, in more heavily polluted regimes, the effects of q_{750} become more pronounced. Figure 4 (c) shows that moister environments support higher LWP values at high N_d. At high q_{750}, the free troposphere can act as a moisture source, whereas under drier conditions, the evaporation-entrainment mechanism depletes the LWP.

In low LTS environments, the LWP decreases with increasing N_d, with stronger decreases seen in drier cases. This is consistent with the evaporation-entrainment mechanism. Figure 4 (f) shows that the difference between the high and low humidity regimes manifests at a lower droplet number than in the high LTS regimes (Figure 4 (c)). This importance of humidity at lower N_d may be due to increased turbulent mixing with the above-cloud at low LTS. This would enhance the rate of droplet evaporation, thereby increasing the dependence of LWP on q_{750}.

When comparing across stability regimes, Figures 4 (g) and (h) show that when the N_d is low, low LTS conditions support clouds with a higher LWP. This is consistent with a deepening of the boundary layer under unstable conditions, allowing clouds to grow deep enough to precipitate, such that an increase in aerosol allows for more LWP to be retained in the cloud through the precipitation suppression mechanism. Additionally, low LTS facilitates the vertical transport of moisture, thereby promoting cloud formation (Kay et al., 2016). This behaviour is similar in both high and low q_{750} environments, suggesting that humidity plays a smaller role under these conditions.

Arctic clouds have previously been observed to have higher LWP values in low LTS environments than in stable ones (Barton et al.; Taylor et al., 2015; Yu et al., 2019). This may be because moisture inversions are common in the Arctic (Solomon et al., 2011), such that mixing with the free troposphere under unstable conditions can augment the LWP. However, Figures 4 (g) and (h) show that this increased LWP at lower LTS does not hold as aerosol load increases. At high N_d, low LTS environments have lower LWPs, with a greater difference at low humidity. The above-cloud moisture is potentially insufficient to offset the increased droplet evaporation in more polluted environments, leading to the shift to lower LWP, particularly in drier environments.

4 Discussion

In this work, we have investigated the factors which influence the N_d-LWP relationship in Arctic clouds. We have found that LTS is a dominant control on the LWP response, with increases in LWP possible in high stability conditions. Specific humidity only appears to influence the relationship in polluted or high LTS conditions, while the background N_d state exerts little control on the LWP response. However, despite careful filtering to remove instances in which the data are prone to errors, some uncertainties in the results remain.

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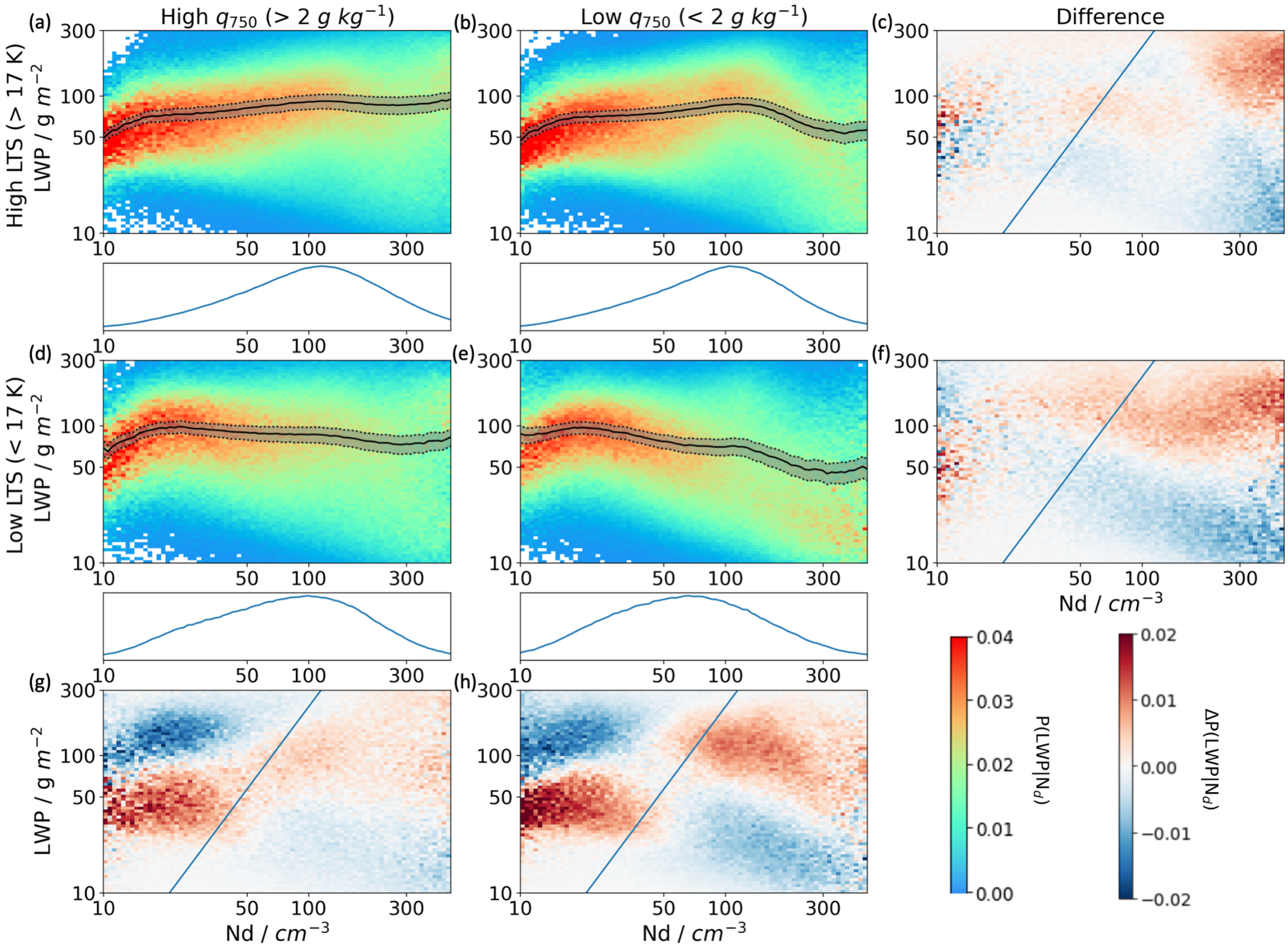

Figure 4. Joint probability histograms for the N_d-LWP divided into four meteorological regimes based on LTS and q. The difference plots are shown at the end of each row and column, with red over blue indicating higher LWP at higher humidity/LTS. The black lines and grey shading on the joint probability histograms represent the mean LWP value for each N_d bin and the 95 % confidence interval, respectively. The blue lines on the difference plots indicate clouds with effective radius of 15 μ m, so clouds to the right of the line are expected to be non-precipitating.

For example, although the pixels have been filtered by cloud top temperature, the misclassification of mixed-phase clouds as liquid may influence the results. However, Khanal and Wang (2018) showed that the error associated with this misclassification is small when compared those generated by high solar zenith angles experienced in the Arctic, which in turn has been addressed in this work by the omission of high-angle pixels. Correlated errors in the LWP and N_d MODIS retrievals may also introduce biases (Gryspeerdt et al., 2019). However, the positive relationship between MODIS N_d and AMSR-E LWP, which would not be affected by correlated errors, suggests that this is a real relationship and not a retrieval artefact.

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Despite ERA5 performing better than other reanalysis datasets when compared to in-situ observations of temperature and humidity (Graham et al., 2019), the meteorological conditions in the Arctic are still poorly constrained. Therefore, use of the reanalysis data may introduce additional uncertainties into the results. However, Renfrew et al. (2021) found that ERA5 compared well to in-situ observations of ice-free regions in the Arctic, so these uncertainties are unlikely to strongly impact the findings of this work.

As the Arctic warms, the LTS is projected to decrease (Boeke et al., 2021). Figure 4 shows the N_d-LWP relationship would become more negative under these conditions. Therefore, the aerosol indirect effect would be weaker due to less strong short-wave cooling for a given increase in N_d.

Equally, these results also show that the LWP response to changes LTS are different in clean and polluted environments. This is key as industrialisation and the creation of new trans-Arctic shipping lanes are projected to be developed as the Arctic heats and sea ice retreats, introducing a large new source of anthropogenic aerosols (Persson, 2012; Schmale et al., 2018). From Figure 4, moving from a clean to a polluted regime in lower LTS environments leads to a decrease in cloud LWP. Therefore, the aerosol cooling effect is weakened in a warmer, more polluted environment.

The results presented here are only for clouds over ocean; more work is required using different datasets to see if they hold in ice-covered regions. Nevertheless, these findings that the aerosol-cloud interactions change with warming and that the LWP-LTS relationship depends on the aerosol loading may have significant implications for the surface energy budget in a rapidly changing Arctic.

5 Conclusions

Previous studies have found a strong sensitivity of Arctic cloud properties and aerosols (Garrett et al., 2004; Coopman et al., 2018). However, these works were either of a limited spatial extent or considered the average response of cloud properties across the Arctic, and therefore did not observe the spatial heterogeneity in cloud response. This work considered the regional variation in the LWP response to N_d, documenting a positive sensitivity at higher latitudes. Positive relationships have previously been observed under some conditions, but not at the strength found in this work (Han et al., 2002; Chen et al., 2014; Toll et al., 2019; Gryspeerdt et al., 2019). However, the response is typically negative across the globe. The seasonal variation in the sensitivity indicates that this signal was most strongly observed during the summer months (Figure 1). Comparison of cloud and meteorological properties of the regions displaying positive and negative sensitivity indicates that stability, in particular LTS, is a significant driving force for the difference in behaviour (Figure 2).

There is only a weak response to cloud-top specific humidity, but the variation with LTS was much greater (Figure 3). Under moist, stable conditions, the LWP increases with N_d, as seen with subtropical clouds (e.g. Chen et al., 2014). Even when considering cases with lower humidity, increases in LWP with N_d are supported up until high N_d, at which point the humidity is insufficient to offset the moisture lost to droplet evaporation (Figure 4). The frequency of these high LTS conditions at high latitudes during the summer months explains the spatial pattern in the sensitivity in Figure 1.

Unstable conditions generate higher LWP values than stable conditions for low N_d, potentially due to precipitation suppression (Figure 4). Therefore, in a future, lower-LTS environment, clouds have a stronger shortwave cooling effect. However, in a more polluted Arctic, the response to low LTS is different; interactions with aerosols would produce lower LWP clouds, thereby reducing the aerosol cooling effect.

These findings on the dependence of the N_d-LWP relationship on the LTS and cloud background state have significant implications for a warmer, more polluted Arctic. Projected increases in aerosol concentrations in conjunction with a decrease in the LTS may ultimately lead to a thinner, lower-LWP clouds, with a reduced cooling potential.

Data availability. The MODIS data were obtained from NASA Goddard Space Flight Centre (https://modis.gsfc.nasa.gov/data/). The ERA5 data were obtained from the Climate Data Store (https://cds.climate.copernicus.eu/). The sea ice and AMSR-E data were obtained from the National Snow and Ice Data Centre (https://nsidc.org/data/).

Author contributions. Both authors contributed to study design and interpretation of results. RMW performed the analysis and prepared the manuscript, with comments from EG.

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

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