# Vertical distribution of microphysical properties of Arctic springtime low-level mixed-phase clouds over the Greenland and Norwegian Seas

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15 Abstract. This study aims to characterize the microphysical and optical properties of ice crystals and 16 supercooled liquid droplets within low-level Arctic mixed-phase clouds (MPCs). We compiled and analyzed 17 cloud in situ measurements from 4 airborne spring campaigns (representing 18 flights and 71 vertical profiles in 18 MPCs) over the Greenland and Norwegian Seas mainly in the vicinity of the Svalbard Archipelago. Cloud phase 19 discrimination and representative vertical profiles of number, size, mass and shape of ice crystals and liquid 20 droplets are established. The results show that the liquid phase dominates the upper part of the MPCs. High 21 concentrations (120 cm<sup>-3</sup> in average) of small droplets (mean values of 15 µm), with an averaged LWC of 0.2 22 gm<sup>-3</sup> are measured at cloud top. The ice phase dominates the microphysical properties in the lower part of the 23 cloud and beneath it, in the precipitation region (mean values of 100 µm, 3 L<sup>-1</sup> and 0.025 g m<sup>-3</sup> for diameter, 24 particle concentration and IWC respectively). The analysis of the ice crystal morphology shows that the majority 25 of ice particles are irregularly shaped or rimed particles, the prevailing regular habits found are stellars and 26 plates. We hypothesize that riming and diffusional growth processes (including the Wegener-Bergeron-Findeisen 27 mechanism) are the main growth mechanisms involved in the observed MPCs. The impact of larger scale 28 meteorological conditions on the vertical profiles of MPC properties was also investigated. Large values of LWC 29 and high concentration of smaller droplets are possibly linked to polluted situations and air masses origins from 30 the South, which lead to very low values of ice crystal size and IWC. On the contrary, clean situations with low 31 temperatures exhibit larger values of ice crystal size and IWC. Several parameterizations relevant for remote 32 sensing or modeling studies are also determined, such as IWC (and LWC) - extinction relationship, ice and 33 liquid integrated water paths, ice concentration and liquid water fraction according to temperature.

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# 36 1. Introduction

The Arctic region is more sensitive to climate change than any other region of the Earth (Solomon et al., 2007).
 Clouds and particularly low-level mixed-phase clouds related processes have a major impact on the Arctic

surface energy budget (Curry, 1995; Curry et al., 1996; Morrison et al., 2011). Observations suggest that
 boundary layer mixed-phase clouds (MPCs, mixture of liquid droplets and ice) are ubiquitous in the Arctic and

40 boundary layer mixed-phase clouds (MPCs, mixture of liquid droplets and ice) are ubiquitous in the Arctic and 41 persist for several days under a variety of meteorological conditions (Mioche et al., 2015; Morrison et al.,

42 2012; Shupe et al., 2011; Shupe and Intrieri, 2004). They occur as single or multiple stratiform layers of

43 supercooled droplets near the cloud top in which ice crystals can form and precipitate. These clouds have a large

44 impact on the surface radiative fluxes and Arctic climate feedbacks (Kay et al., 2012; Kay and Gettelman,

45 2009). The strong impact of MPCs on the energy budget stems from their persistence and microphysical

46 properties which result from a complex web of interactions between numerous local and larger scale processes

47 that greatly complicate their understanding and modeling (Klein et al., 2009; Morrison et al., 2012).

48 However, major uncertainties limit our understanding of the interactions and feedbacks between the physical

49 processes involved in their life cycle. This complexity translates in the large discrepancies that can be found in 50 numerical models to represent the cloud processes, which in turn impacts their capability to forecast cloud

51 properties in the Arctic. For instance, Global Climate Models (GCM) tend to underestimate the amount of liquid

52 water in MPCs (Komurcu et al., 2014). Therefore, the representation of ice formation and growth processes and

53 their interactions with the liquid phase (e.g. liquid/ice partitioning, Wegener-Bergeron-Findeisen process) has to

54 be improved, as already shown in previous modelling studies (**Prenni et al. (2007**) or **Klein et al. (2009**)).

55 Among the various cloud properties which need to be described more accurately, the cloud thermodynamic

56 phase is a parameter of primary importance since it governs the cloud optical and therefore radiative properties

57 as well as its life cycle (longevity and precipitation formation).

However, measuring the spatial phase distribution in low level Arctic mixed-phase clouds, in order to relate it to environmental conditions (height, temperature, surface conditions, air mass origins...) to parameterize and model it, remains a challenge. The parameterizations of liquid and ice partitioning in numerical simulations vary from one model to another. A study carried out by **Klein et al.** (2009) compared outputs from 26 different numerical models. They found that using different schemes of temperature dependent partitioning yield to liquid

 $63 \qquad \text{water content ranging from 12 \% to 83 \% for a same cloud top temperature of -15°C}.$ 

64 Beyond the experimental limitations related to the accurate measurement of the phase partitioning (discussed 65 hereafter), the cloud phase quantification is also hampered by difficulties to translate observational 66 characterization into realistic representations for cloud models with a wide range of scales. The definition of 67 mixed-phase system is actually controversial. A mixed-phase cloud can be regarded as a complete cloud system 68 that contains both liquid and ice involved in mixed microphysical processes but does not necessarily implies that 69 all volumes in the system contain both phases (Shupe et al., 2008). Additionally, the definition of a mixed-phase 70 cloud or volume could be based either on a threshold value for its optical properties or for the ratio between 71 supercooled liquid droplets and ice crystal mass or number (Cober et al., 2001). The threshold values are 72 questionable. The standard assumption in climate models is that liquid and ice are uniformly mixed throughout 73 each entire model grid box (with a typical horizontal resolution of 100 km and 1 km in the vertical, Tan and 74 Storelymo (2016)). However some field measurements (see among others Rangno and Hobbs (2001) or 75 Korolev and Isaac (2003)) suggest that different pockets of solely water or ice in mixed-phase regions coexist

76 with typical scale of tens of meters. This has consequences on how processes like the Wegener-Bergeron-

- 77 Findeisen process should be parameterized in large scale models.
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80 A better assessment of the ice/liquid partitioning will improve our understanding of the life cycle and more 81 precisely the persistence of MPCs since modelling studies show that this persistence is governed by a delicate 82 balance between dynamical, radiative and microphysical processes occurring mainly in the boundary layer 83 (Savre and Ekman, 2015). This understanding is still limited by the description of the microphysical processes 84 related to the initiation and the maintenance of the ice phase. The cloud processes responsible for the production 85 of ice crystals in the upper part of the cloud seems to be mostly driven by the cloud top temperature and the 86 entrainment rates (Savre and Ekman, 2015). In particular, the assessment of IN concentration and its time 87 evolution is of primary importance rely on a very limited set of *in situ* observations and need to be improved 88 (Ovchinnikov et al., 2014). The ice crystal number concentrations usually exceed the number of IN particles. 89 These discrepancies could be explained by the limitations of *in situ* instruments and especially the 90 overestimation of the ice crystal number due to the shattering of large ice crystals on the probe inlets or the 91 inability of instrument measuring IN particles to detect all the activation modes (Baumgardner et al., 2012; 92 Korolev et al., 2011). Secondary ice formation processes or the recycling of IN particles through subcloud 93 sublimation (Lawson et al., 2001; Rangno and Hobbs, 2001; Solomon et al., 2015) may also play an important 94 role and explain such discrepancies. Given the temperatures observed in MPCs, heterogeneous ice nucleation 95 mechanisms are preferentially involved. The concentration of large ice crystals (> 100  $\mu$ m) in particular may be 96 due to heterogeneous ice formation mechanisms (Eidhammer et al., 2010; Prenni et al., 2009). However which 97 process, among deposition, condensation, immersion or contact freezing processes, is mainly responsible for the 98 initiation of ice crystals is still under debate as modeling studies fail to reproduce the observed ice number 99 concentration (Avramov and Harrington, 2010; Fridlind et al., 2007). In a recent modeling study linked to the 100 Aerosol-Cloud Coupling and Climate Interactions in the Arctic (ACCACIA) campaign, Young et al. (2017) 101 showed that small differences in the predicted ice concentration can have large effects on the microphysical 102 structure (such as ice/liquid partitioning) and life time of single layer MPCs. They suggested that the method of 103 parameterizing primary ice concentration in bulk microphysical models is therefore of primary importance.

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105 The recent developments of ground based stations (Barrow, EUREKA, NY-Alesund among others) and 106 spaceborne remote sensing observations (as lidar and radar observations from the CALIPSO and CloudSat 107 platforms respectively) allow today reliable studies of Arctic cloud phase variability from a few km to the pan-108 arctic region (Dong et al., 2010; Kay and Gettelman, 2009; Liu et al., 2012; Shupe et al., 2011). Moreover, 109 remote sensing observations from space performed by active instruments onboard CALIPSO (Winker et al., 110 2003) and CloudSat (Stephens et al., 2002) satellites as a part of the A-Train constellation provide a unique way 111 of characterizing Arctic cloud vertical properties. However, the cloud phase distribution and characterization are

112 highly dependent of the measurement principle of the instruments.

113 The aforementioned techniques provide cloud properties typically averaged over one kilometer, which may be 114 insufficient to study cloud processes at a microphysical scale (i.e. measurements of microphysical cloud

115 properties, spatial resolution less or equal to 100 m). In situ observations are based on direct measurement

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- techniques at a higher spatial resolution (generally < 100 m). Numerous previous studies dedicated to the
- 117 assessment of the *in situ* microphysical properties of Arctic clouds focused on specific case studies (Avramov et
- 118 al., 2011; Gayet et al., 2009; Rangno and Hobbs, 2001; Verlinde et al., 2007). Statistical analysis of mixed-

119 phase cloud properties derived from several *in situ* datasets or airborne campaigns are very scarce and often

focus on the Western Arctic region (McFarquhar et al., 2007; Jackson et al., 2012). Such data analysis strategy is still missing in the European Arctic Region (and in the vicinity of Svalbard, over the Greenland and Norwegian Seas in particular).

In Mioche et al. (2015), the spatial, seasonal, and surface conditions variability of MPC properties using CloudSat and CALIPSO spaceborne observations has been investigated. The study showed a large occurrence of MPCs all year long both over the whole Arctic and the Svalbard regions. It was clearly evidenced that the Svalbard region, due to its specific location near the Atlantic Ocean, presents a larger occurrence of low level MPCs compared to the averaged Arctic. Then, it appears important to investigate the microphysical properties of MPCs in the Svalbard / Greenland Sea regions from a statistical point of view to provide representative profiles that can be compared to previous works focused on the Western Arctic region.

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131 This work provides statistics analysis of liquid and ice properties of low level Arctic MPCs from in situ data 132 collected in single layer MPCs during several airborne campaigns in the region of Norwegian/Greenland Sea 133 carried out between 2004 and 2010. We compiled observations of microphysical composition of Arctic mixed-134 phase clouds (cloud phase, hydrometeors number, mass and shape) to present vertical profiles of liquid and ice 135 properties. The main objective is a step to a better understanding of the processes involved in Arctic low-level 136 MPC life cycle at the microphysical scale. We aimed to relate these properties to environmental conditions in 137 order to improve the cloud parameterizations used in models and remote sensing algorithms. The results will 138 also complement previous works concerning Arctic clouds characterizations performed in Western Arctic.

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140 This paper is organized in four sections. The description of the field experiments, instrumentation and datasets 141 will be made in section 2. Section 3 will present and discuss the vertical profiles of microphysical properties of 142 the low-level MPCs. Finally, key parameterizations useful for modeling or remote sensing will be proposed in 143 section 4.

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#### 2.1 Airborne campaigns

This study is based on *in situ* data collected in single-layer mixed-phase clouds (MPCs) during four international
airborne campaigns organized in the "European" Arctic region, namely:

Field experiments, airborne measurements and meteorological situations

(i) and (ii) the Arctic Study of Tropospheric Aerosols, clouds and Radiation experiments (ASTAR, Herber et al., 2004; Jourdan et al., 2010; Ehrlich et al., 2009; Gayet et al., 2009; Lampert et al., 2009) which took
place in the vicinity of Svalbard (Longyearbyen, Norway, 78° N, 15° E) in April 2004 and April 2007. The
Polar-2 aircraft operated by AWI (Alfred Wegener Institute) was flown during these two experiments;

154 (iii) the Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate, Chemistry,

Aerosols, and Transport (POLARCAT-France, **Delanoë et al., 2013; Law et al., 2008; Quennehen et al., 2011**)

156 which was carried out in northern Sweden (Kiruna, 68° N, 20° E) in April 2008 during the International Polar

157 Year. Measurements were performed onboard the French ATR-42 aircraft of SAFIRE (Service des Avions

158 Français Instrumentés pour la Recherche en Environnement);

- and (iv) the Solar Radiation and Phase Discrimination of Arctic Clouds experiment (SORPIC, Bierwirth et al.,
- 160 **2013**), in the Svalbard region in May 2010 with the AWI Polar-5 aircraft.
- 161 All the clouds sampled during these four campaigns were located over the Arctic Greenland and Norwegian seas
- as displayed on Fig. 1. The scientific flights during ASTAR and SORPIC covered latitudes ranging from 75° N
- 163 to  $79^{\circ}$  N (Greenland Sea) while the flights during POLARCAT were performed between  $70^{\circ}$  N and  $73^{\circ}$  N
- 164 (Norwegian Sea). Moreover, the data were all collected during spring (April and May).

For this study, we restricted the measurements to continuous ascent and descent flight sequences into singlelayer MPCs at the aircraft speed (between 80m/s and 100 m/s for all campaigns) since our main objective is to study the vertical partitioning of ice and liquid thermodynamical phases. Our dataset consists of 71 cloud profiles (see Table 1) representing more than 21 000 measurement points at 1Hz (350 minutes of cloud observations), spread out over 18 flights performed above arctic open sea water.

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# 2.2 In situ instrumentation

A similar *in situ* instrumentation was loaded on the three aircraft: the German Polar-2 and Polar-5 and the French ATR-42. The same data processing procedure was used in order to derive the cloud microphysical parameters (at a same scale: i.e. ~ 100 m). This consistent cloud data set is used to achieve a statistically representative description of the properties of Arctic mixed-phase clouds sampled over the Greenland and Norwegian Seas during spring.

- 178 The suite of *in situ* instruments used to measure the MPC microphysical and optical properties consists of:
- 179 the Cloud Particle Imager (CPI, Lawson et al., 2001), which captures cloud particle images on a 180 1024x1024 pixels CCD camera with a pixel resolution of 2.3 µm and with 256 grey levels. At least 5 181 pixels are necessary to identify a cloud particle, so the particle sizes derived from the CPI range from 15 182 um to approximately 2 mm. The images are processed using the software developed at the Laboratoire 183 de Météorologie Physique (LaMP, Lefèvre, 2007) based on the original CPIView software (CPIView, 184 2005, Lawson et al., 2001; Baker and Lawson, 2006). In particular, it provides particle size 185 distribution (PSD) and derived microphysical parameters such as particle concentration, effective 186 diameter, extinction coefficient and ice water content as well as a particle habit classification. The data 187 processing method used to derive the extinction coefficient ( $\sigma$ ) and the Ice Water Content (IWC) is 188 described in Appendix A;
- the PMS Forward Scattering Spectrometer Probe (FSSP-100, Baumgardner et al., 2002; Knollenberg, 190
   1981); it provides the droplet size distribution from 3 to 45 μm. The derived parameters from the PSD are the droplet concentration, the effective diameter, the extinction coefficient (σ) and the liquid water content (LWC);
- the Polar Nephelometer (PN, Gayet et al., 1997) which measures the angular scattering intensities (non normalized scattering phase function) of an ensemble of cloud particles (either droplets, ice crystals or a mix), from a few micrometers to about 800 μm. In particular, these measurements are used to identify spherical from non-spherical particles and thus discriminate the dominant cloud thermodynamical

phase. The extinction coefficient and the asymmetry parameter (g) are calculated following the methodology presented in **Gerber et al. (2000)** and **Gayet et al. (2002)**;

- the Nevzorov probe (Korolev et al., 1998), which uses the hot-wire technique to retrieve the liquid
   water content and the total water content. Note that the Nevzorov data are only used to determine liquid
   water content during ASTAR 2004 because the FSSP-100 was not used during this campaign. The
   retrieval method used to determine the liquid water content is described in Appendix A.
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204 All these cloud probes were heated in order to avoid icing during the flights. The combination of these probes 205 provides the microphysical properties of cloud particles from a few micrometers (typically 3 µm) to about 2 mm. 206 Data are recorded at 1 Hz frequency which corresponds to a spatial resolution of about 100 m (according to the 207 aircraft speed). The uncertainties and measurement ranges associated to the derived cloud parameters are 208 summarized in Table 2. However it should be noted that *in situ* measurements accuracy may be hampered by the 209 shattering of large ice crystals on the probe inlets, inducing smaller particle artifact (Heymsfield, 2007) leading 210 to an overestimation of small particle concentration. For example, previous studies of Field et al. (2003) and 211 Heymsfield (2007) showed that shattering effect may lead to an overestimation of about 20 % on the bulk 212 properties and a factor 2 or 3 on the number concentration of ice crystals. Moreover, the recent study by Guyot 213 et al. (2015) that compared on *in situ* measurements with the same probes and similar inlet design in a wind 214 tunnel experiment showed that measured particles can vary from one instrument to another and careful 215 calibration is needed. Even through no standard method was available during the campaigns to accurately 216 determine and remove the impact of shattering (designed tips, particle interarrival time measurement...), a short 217 analysis is described in Appendix B for the control of the data quality and the significance of shattering effect. 218 Comparing the extinction coefficient measured by the PN measurements to the extinction derived from the 219 combination of the CPI and FSSP measurements showed that the shattering effect, in our case, was smaller than 220 the measurements uncertainties (i.e. 25 %, 35 % and 55 % for PN, FSSP and CPI respectively see Table 2).

The three research aircraft also measured basic meteorological parameters along the flight track (see Gayet et al., 2009). We recall that the static air temperature is calculated with an accuracy better than  $\pm$  0.5 K. If high liquid water contents can alter these temperature measurements, the observed contents, lower than 0.6 g m<sup>-3</sup> during most of the MPC flights, ensure that this effect was not significant along the cloud transects. The altitude and geographical position parameters were measured by the airborne GPS systems with an accuracy of 50 m.

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#### 2.3. Normalized altitudes

229 Table 3 summarizes, for the 71 selected profiles, the statistics of altitudes for MPCs top and base, as well as the 230 thickness of the cloud layer containing liquid water. The mean cloud top altitude is located around  $1200 \pm 310$  m 231 while the mean cloud base altitude (referring as the altitude below which liquid phase is no longer present) is 756 232  $\pm$  283 m. This is consistent with observations performed in western Arctic where cloud top altitudes lie between 233 885 m and 1320 m, and cloud base altitudes between 420 m and 745 m (McFarquhar et al., 2007). Our 234 measurements also indicate that the thickness of the liquid layer spans from 100 m to 950 m with an averaged 235 value of 444 m. The objective of this study is to merge and analyze the MPC microphysical data obtained during 236 the four airborne campaigns to derive representative vertical profiles. Since cloud top and cloud base heights

exhibit large variability (see Table 3), the altitudes are normalized following the method presented in **Jackson et** al. (2012). The cloud top and cloud base refer to the liquid phase layer, i.e. the cloud layers containing liquid droplets (mixed-phase or liquid only). These layers are identified based on the PN asymmetry parameter values greater than 0.8 (**Jourdan et al. (2010**), see section 2.4. below). Within these layers (Eq. (1)) and below the cloud base (Eq. (2)) the normalized altitudes  $Z_n$  are defined as follow:

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$$\boldsymbol{Z}_{\boldsymbol{n}} = \frac{\boldsymbol{z} - \boldsymbol{z}_{b}}{\boldsymbol{z}_{t} - \boldsymbol{z}_{b}} \qquad \qquad \text{for } \boldsymbol{z}_{b} < \boldsymbol{z} < \boldsymbol{z}_{t} \tag{1}$$

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$$245 \qquad \mathbf{Z}_n = \frac{\mathbf{z}}{\mathbf{z}_b} - \mathbf{1} \qquad \text{for } \mathbf{z} < \mathbf{z}_b \tag{2}$$

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where  $Z_n$  is the normalized altitude, Z the altitude corresponding to the aircraft measurements,  $Z_t$  and  $Z_b$  the cloud top and base altitudes respectively. Thus, an altitude of 1 corresponds to the top of the cloud liquid containing layer and 0 to its base. Negative values characterize regions of ice precipitation below the cloud layer and the altitude of -1 defines the ground level according to Eq. (2).

To obtain representative statistical results, the cloud layers have been stratified in 10 levels with intervals of 0.2

of normalized altitude, each containing around 2000 observations (i.e. about 10% of the data set). The vertical profiles of MPC microphysical properties presented hereafter are obtained by averaging the *in situ* measurements over each normalized altitude layer. The profiles are computed for the whole data set and for each main meteorology situation separately (see section 2.5) for a better analysis and discussions of the results.

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## 2.4. Determination of the cloud thermodynamical phase from *in situ* measurements

259 As stated above, the asymmetry parameter (g) derived from the PN scattering phase function (PhF) 260 measurements is used to discriminate cloud thermodynamic phase. Indeed, in a previous study, Jourdan et al. 261 (2010) have shown with a principal component analysis that g is a reliable proxy to determine the cloud phase of 262 Arctic MPCs. One can notice that our phase discrimination is considered from an optical point of view and 263 differs from the one used by Korolev et al. (2003) which is based on the ice water fraction (IWC/TWC) to 264 identify cloud phase. Large values of g > 0.83 are typical of an ensemble of particles optically dominated by 265 liquid water droplets where ice crystals do not significantly affect the optical properties. On the contrary, smaller 266 values of g (< 0.80) are characteristic of a cloud optically dominated by ice crystals, with negligible contribution 267 of liquid droplets. For g ranging from 0.80 to 0.83, both liquid droplets and ice crystals contribute to the optical 268 properties. The optical signature of the ice is more pronounced (i.e. g decreases) as the concentration and/or the 269 size of ice particles becomes larger. These results are well illustrated and discussed by Febvre et al. (2012) 270 where PN measurements were combined with FSSP and CPI data.

Figure 2 displays the mean PN scattering phase function (Fig. 2a) according to the normalized MPC altitude levels as well as the vertical profile of the corresponding g-values (Fig. 2b). At cloud top, the PhF is characterized by a rather high scattering at forward angles (angles lower than  $60^{\circ}$ ) associated to lower scattering at sideward angles ( $60-130^{\circ}$ ), and enhanced scattering around  $140^{\circ}$ . These features are representative of cloud layers dominated by spherical particles (mainly supercooled liquid droplets), corresponding to typical g-values greater than 0.83. As Z<sub>n</sub> decreases, the PhF becomes smoother and more featureless. A side scattering 277 enhancement is observed along with an attenuation of the 140° peak. This behavior can be attributed to the 278 presence of non-spherical ice crystals increasing towards the cloud base. This is in agreement with the 279 continuous decrease of g-values observed from cloud top (0.84) to cloud base (0.82). Figure 2 also shows that 280 the ice phase region below the cloud layer (-1 < Zn < 0) is characterized by a featureless and flat (at side 281 scattering angles) PhF, with no significant influence of the altitude. These PhF are associated to g-values smaller 282 than 0.8. It is thus clearly shown that the PhF and asymmetry parameter are related to specific microphysical 283 properties encountered at different cloud levels. These observations demonstrate that the PhF and the asymmetry 284 parameter can be regarded as an accurate signature of the main microphysical properties observed in the MPC 285 layers particles.

The liquid droplets properties are determined from the FSSP or Nevzorov probe measurements when g-values are greater than 0.8 (i.e. indicating a "liquid-containing" phase). Accordingly, the ice crystals properties are derived from CPI measurements when g-values are lower than 0.83 (i.e. indicating an "ice-containing" phase). For g ranging between 0.8 and 0.83, both liquid and ice properties are derived. Moreover, CPI particle images classified as spherical droplets are not taken into account for the determination of ice crystal microphysical parameters. Table 4 summarizes the cloud phase analysis.

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#### 2.5. Meteorological situations

296 All the selected situations correspond to low-level single layer mixed-phase clouds in the boundary layer during 297 spring. If these criteria ensure the homogeneity of the dataset, weather conditions still vary significantly from 298 one campaign to another or even within a campaign. In order to provide a comprehensive dataset to improve 299 model parameterization, this is of great importance to discriminate and classify the observations depending on 300 environmental conditions. The most trivial classification is the temperature regime. Savre and Ekman (2015), 301 showed that it is one of the major factors (with cloud top entrainment) controlling the production of new ice 302 crystals and the maintenance of MPCs. In the present study, two temperature regimes have been selected based on the mean cloud top temperature of each situation: the "cold" situations (-22  $^{\circ}C < T_{Top} < -15^{\circ}C$ ) and the 303 304 "warm" situations (-15 °C < T<sub>Top</sub> < -8°C). In spring, the cold polar vortex that covers the Arctic Region weakens 305 and inclusions of midlatitude air masses are more likely. Atmospheric properties such as temperature, humidity, 306 and particles loading can change significantly. Arctic cloud properties are then strongly linked to the air mass 307 origin (Gultepe et al., 2000; Gultepe and Isaac, 2002), since their formation is driven by the aerosol particles 308 properties and thermodynamical and dynamical conditions. For these reasons, we also included the air mass 309 origin in the classification, which was determined from the analysis of back-trajectories computed with the 310 NOAA HySPLIT model (Hybrid Single-Particle Lagrangian Integrated Trajectory model, Draxler and Rolph, 311 2003). The back trajectories can be classified in two kinds: the air masses originating from the North (over sea 312 ice or open water of the Arctic Ocean and Greenland Sea) that are cold and clean, and the air masses more 313 continental which have travelled over more polluted regions in the South and/or East. We made the choice of 314 only two temperature regimes and two main air mass origin in order to ensure representative and statistically 315 significant datasets for each class. Table 5 shows that all the cold situations are correlated with a north origin air 316 mass (blue in Table 5). Among the 12 warm situations, 7 correspond to air masses originating from North (green

- 317 in Table 5) and 5 from South/East (red in Table 5). So, at the end, this classification leads to only 3 types of
- 318 situations: (i) Cold cloud top temperature situations with air masses originating always from the North (hereafter
- 319 referred as COLD in the manuscript); Warmer situations with air masses which has its origin either (ii) in the
- 320 North (hereafter WARM\_NO cases) or (iii) from the continent: South and East (hereafter WARM\_SO cases).
- 321 The mean vertical profiles of temperature of these 3 regimes are displayed on Fig. 3. The results show a well
- 322 pronounced temperature inversion (~ -10 °C) for the WARM\_SO situations whereas WARM\_NO cases do not
- 323 exhibit such clear temperature inversion. The COLD situations are characterized by a temperature of -20 °C at
- 324 cloud top and by weather situations dominated by cold air outbreaks from higher latitudes (Gayet et al., 2009).
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#### 26 3. Vertical properties of liquid droplets and ice crystal particles within MPCs

The purpose of this section is to provide a quantitative assessment of the average microphysical and optical properties of the MPC cloud layers at a spatial scale of approximately 100 m. The vertical profiles presented in this study come from aircraft *in situ* measurements and are obtained from several distinctive clouds. It should be emphasized that these profiles cannot be strictly regarded as vertical and instantaneous profiles (each ascending or descending flight sequence is generally made in 5 to 10 min). It differs from the remote sensing measurements that usually provide snapshots of a same cloud.

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# 3.1. Liquid phase properties

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337 Figure 4 displays the average vertical profiles expressed with the normalized altitude reference for the liquid 338 phase properties: the extinction coefficient, the droplet number concentration, the liquid water content and the 339 effective diameter (Figs. 4a to 4d). These profiles are obtained using FSSP-100 or Nevzorov probe 340 measurements and constrained by PN g-values greater than 0.8. On this figure, the average profiles for liquid 341 properties are discriminated for each environmental conditions class: COLD in blue, WARM NO in green and 342 WARM SO in red. The mean profile corresponding to the average over all situations (or campaigns) is also 343 shown (in black). The average vertical distribution for the liquid droplet number size distribution is shown in 344 Fig. 4e.

345 The MPC properties are characterized by increasing values of LWC with altitude. LWC mean values range 346 between 0.1 g m<sup>-3</sup> at the bottom of the liquid layer and nearly 0.2 g m<sup>-3</sup> close to the cloud top. The concentration 347 of cloud droplets remains nearly constant throughout the MPC layers with mean values around 120 cm<sup>-3</sup>. 348 However smaller values are observed near the cloud top. Clouds corresponding to the WARM SO situations are 349 characterized by larger values of droplet concentration and LWC (200 cm<sup>-3</sup> and 0.3 g m<sup>-3</sup>) compared to the 350 COLD and WARM NO cases. This is related to the fact that air masses originating from midlatitudes are more 351 humid. The extinction coefficient profiles are correlated with the LWC measurements indicating that liquid 352 droplets mainly drive the optical properties of the upper MPC layers. This is consistent with the observed shape 353 of the scattering phase function at cloud top displayed in Figure 2. The extinction coefficient presents maximum 354 values in the upper part of the cloud (average around 25 km<sup>-1</sup>), and smaller extinction in the lower part of the 355 liquid layer (down to 15 km<sup>-1</sup>). Finally, the vertical profiles of the effective diameter (Fig. 4a) and droplet 356 number size distribution (Fig. 4e) are consistent with the extinction coefficient, LWC and droplet concentration.

- 357 Indeed, the effective diameter is proportional to the ratio of the LWC to the extinction coefficient. The cloud
- 358 layers dominated by the liquid phase exhibit small droplet sizes, with a slight increase of the diameter from cloud
- base to cloud top (from 10 to 15  $\mu$ m).
- 360 The main features of the vertical distribution for the liquid phase properties are in agreement with previous
- observations (e.g. Lawson et al. (2001), McFarquhar et al. (2007) or Jackson et al. (2012)). These studies
   focused on MPCs in the western Arctic region under meteorological situations that can be connected to the ones
- 363 presented in our work. Lawson et al. (2001) studied a boundary layer MPC in spring over the Beaufort Sea
- 364 during the First International Satellite Cloud Climatology Regional Experiment Arctic Cloud Experiment (FIRE-
- ACE). The temperature range lied between -22 °C and -25 °C. This case could be regarded as the COLD
- situations in our study. Lawson et al. (2001) showed LWC values around 0.15 g m<sup>-3</sup> and droplet concentration
   close to 200 cm<sup>-3</sup>.
- 368 McFarquhar et al. (2007) merged 4 MPC situations (corresponding to 53 cloud profiles) in autumn over 369 Barrow and Oliktok Point, Alaska during the Mixed-Phase Arctic Cloud Experiment (M-PACE). The MPCs 370 were associated with a low-level northeasterly flow over the ice pack resulting in persistent roll clouds at low-371 level altitude. Cloud top temperatures lied between -12 °C and -16 °C. These situations can be related to our 372 COLD and WARM NO cases. They also observed the increase of liquid droplet size, LWC and number 373 concentration with the altitude. The LWC range (0.15-0.19 g m<sup>-3</sup>) is consistent with our study but the droplet size 374 range (from 14 µm at cloud base to 22 µm at cloud top) is slightly larger and the droplet number concentrations 375 significantly lower (between 23 and 72 cm<sup>-3</sup>). Finally, Jackson et al. (2012) merged 41 MPC profiles during the 376 Indirect and Semi-Direct Aerosol Campaign (ISDAC). They observed liquid droplet properties with mean LWC 377 around 0.15 g m<sup>-3</sup> at cloud top, droplet size from 8 to 16  $\mu$ m and droplet concentration around 150 cm<sup>-3</sup>. These 378 results are consistent with our observations of the liquid phase within MPCs observed over the Greenland and 379 Norwegian Seas.
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#### **3.2.** Ice phase properties

383 The ice crystal properties derived from the CPI measurements when the PN g-values are lower than 0.83 are 384 displayed on Figure 5 with the same representation as the one used for the liquid phase. In the following, the ice 385 crystal concentration corresponds to particles larger than 100 µm in order to minimize the effect of potential 386 shattering artifacts on this parameter (see Febvre et al., 2012). However, the extinction coefficient, the ice water 387 content, the effective diameter and particle size distribution are determined using all CPI images excluding those 388 identified as liquid droplets. This choice has been made to be consistent with previous studies and allow for 389 accurate comparisons with microphysical parameters obtained during Western Arctic campaigns. Averaged 390 values of ice crystal concentration (N<sub>i</sub>) and extinction coefficient ( $\sigma_i$ ) are around 3 L<sup>-1</sup> and 0.4 km<sup>-1</sup> respectively. 391 IWC and effective diameter ( $D_{eff,i}$ ) display mean values ranging from 0.01 to 0.035 g m<sup>-3</sup> and from 80 to 130  $\mu$ m 392 respectively. No clear trend on the mean profiles of these properties is observed as no significant correlation with 393 height is found. However, the values of these parameters decrease to nearly zero at cloud top ( $Z_n=1$ ). This 394 indicates that the cloud top layer is almost exclusively composed of supercooled liquid droplets and eventually a 395 very low concentration of small ice crystals as shown by the PSD on Fig 5e. These results corroborate the 396 findings from the previous experiments such as the ISDAC and M-PACE campaigns in Western Arctic

- 397 (McFarquhar et al., 2011; Jackson et al., 2012). These studies were based on 53 cloud profiles during the M 398 PACE campaign (McFarquhar et al., 2011) and 41 cloud profiles during the ISDAC campaign (Jackson et al.,
- 399 2012). The ice crystal properties of single layer MPCs observed over the Beaufort Sea region did not show any
   400 significant vertical variability.
- 401 Typical IWC and particle concentration (for crystals with size larger than  $125\mu$ m) values lied between 0.006 and 402 0.025 g m<sup>-3</sup> and between 1.6 L<sup>-1</sup> and 5.6 L<sup>-1</sup> for the M-PACE situations. These values are similar to those of the
- 403 COLD and WARM\_NO cases of the present study. Averaged values of IWC and particle concentration during
- $404 \qquad \text{ISDAC are in the range of the WARM\_SO situations of the present work with values around 0.02 \text{ g m}^{-3} \text{ and } 0.27$
- 405 L<sup>-1</sup> respectively for the ISDAC situations. The average ice crystal size observed during M-PACE is around 50
- 406 μm which is smaller than the typical size found in our study. It could be explained by less efficient WBF and407 riming processes and smaller droplet number also observed during M-PACE.
- 408 Deeper in the precipitation layer, closer to the sea level (Zn < -0.5) no general trend can be depicted as ice crystal 409 properties show a large variability. Yet, no ice crystals were found in this region for the WARM SO situations,
- 410 whereas the ice precipitation reaches the surface  $(Z_n = -1)$  for the COLD and WARM\_NO regimes.
- 411 The particle shape vertical distribution was also investigated based on the CPI images. It can provide an insight
- 412 of the main microphysical growth processes occurring in such MPCs. Figure 6 displays the particle shape
- $413 \qquad \text{distributions relative to number and mass concentration with $Z_n$ (Figs. 6a and 6b) and temperature (Figs. 6c and 6b) and figs. 6c and 6b) and 6b)$
- 414 6d). To this purpose, particle shapes have been automatically classified by the algorithm developed at LaMP (see
- 415 details in Lefèvre, 2007). In addition, the resulting classification was supported by an accurate human-eye 416 visualization in order to control the results and avoid the main shortcomings linked to the automatic 417 classification. As indicated above, only particles with size greater than 100 µm were taken into account in order 419
- 418 to avoid misclassification of smaller particles and shattering artifacts.
- 419 Our results clearly show that rimed and irregular ice crystals are dominant within MPCs (up to 80 % of the
- total). In particular, irregular ice particles are encountered at all altitude and temperature. They account for 30 %
- 421 to 50 % of the total number concentration (and between 20 % and 30 % of mass concentration) depending on the
- 422 altitude or temperature of the MPC layer. Rimed particles are predominant inside the liquid containing cloud
- 423 layer  $(0 < Z_n < 1)$  with a contribution up to 40 % in number (60 % in mass) where low temperatures (below -18 424 °C) are observed.
- 425 An interesting feature is the significant occurrence (around 40 %) of ice crystals with a predominant a-axis
- 426 growth at all cloud levels. Indeed, plates, sideplanes and stellars are the dominant habits among the regular 427 shapes regardless of the cloud layer altitude.
- 428 Below the cloud ( $Z_n < 0$ ), precipitating ice crystals are characterized by a mass concentration dominated by 429 rimed particles and by large number concentration fraction of irregular ice crystals.
- 430 Over all, these results agree with the ones presented in McFarquhar et al. (2007) based on *in situ* observations
- 431 of MPCs during the M-PACE experiment. McFarquhar et al. (2007) also stated that small supercooled water
- 432 droplets dominated the upper layer of the cloud while larger ice particles were present in the lower part and
- 433 below the cloud (including irregular, aggregate or rimed-branched crystals). But our results differ since they
- 434 observed a fraction of needles and columns particles a lot larger than in our study (resp. up to 50% below the
- 435 cloud versus less than 10 %). On the opposite, our results are not in agreement with the observations described in
- 436 Korolev et al. (1999), this is because they observed even less regular ice crystals: irregular shaped ice crystals

437 accounted for up to 98 % of the total number of ice particles. This disagreement could be explained by two 438 reasons. First, Korolev et al. (1999) considered a wide variety of clouds sampled in the Canadian and US Arctic 439 (stratocumulus and cirrus at temperatures ranging from 0 to -45 °C and up to 7.5 km of altitude) whereas the 440 present study focuses only on MPCs in the Svalbard region at low altitudes. The disagreement may also stem 441 from the different image processing used in these studies. For instance, Korolev et al. (1999) took into account 442 particles larger than 40 µm (while a 100 µm threshold was used in our study) and two ice crystal shapes: pristine 443 (defined as faceted ice single particles) and irregulars were considered (while 10 particles shapes were accounted 444 for in our results).

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#### **3.3.** Discussion on statistical vertical profiles

448 The quantitative estimates of the separate properties of droplets and ice crystals may give an insight on the 449 microphysical processes occurring in MPCs. These processes are involved in the MPC life cycle, in particular to 450 maintain the coexistence of liquid droplets and ice crystals, leading to its persistence (Morrison et al., 2012). 451 More specifically, the increase with height of droplet size and LWC observed in the vertical profiles is consistent 452 with a condensational growth process. The slight decrease on LWC and number concentration observed at the 453 very top of the cloud may be due to turbulent mixing (Korolev et al., 2015) and entrainment of dry air. 454 Additionally, the data collected in this part of the cloud may also lead to a slight underestimation of the LWC 455 since a mixing of cloudy and cloud free patches could be averaged together given the sampling resolution (i.e. 456 100 m). The analysis of the vertical profiles of ice properties and ice crystal shapes (cf. Fig. 6) shows that the 457 presence of pristine particles, mainly plates and stellars could be linked to a very fast ice crystal growth by vapor 458 deposition due to Wegener-Bergeron-Findeisen process (WBF, Bergeron, 1935; Findeisen, 1938; Wegener, 459 **1911**) in which ice crystals grow at the expense of liquid droplets. The large contribution of rimed particles 460 confirmed that riming process shall be significant in a mixed-phase cloud. The prevalence of irregular particles is 461 in agreement with the previous studies from Korolev et al. (1999) and McFarquhar et al. (2007) and suggests 462 that aggregation growth processes, or a combination of several growth mechanisms are involved. This also 463 indicates that turbulence or mixing into the cloud may have an important influence by redistributing the 464 precipitating ice crystals in the upper cloud levels. Measurements of the vertical wind speed (which are not 465 available for these campaigns) would be helpful to confirm this hypothesis.

466 Theoretical adiabatic LWC has also been determined assuming a non-entraining parcel of moist air rising and 467 reaching saturation. It is calculated from the pressure and temperature measurements from cloud base to cloud 468 top. These theoretical values are then compared to the observed LWC values to evaluate the influence of 469 turbulence or mixing effects on LWC as well as the efficiency of ice growth by WBF process or riming 470 processes. The profiles of the adiabatic ratio (the ratio of the adiabatic LWC to the observed LWC) are displayed 471 on Fig. 7. Subadiabatic values are found for all meteorological regimes. This means that processes responsible 472 for a decrease of LWC compared to the adiabatic prediction are prevalent. In particular, this strengthens the 473 assumption that a turbulent entrainment of dry air, resulting in the evaporation of liquid droplets, may occur at 474 cloud top. Moreover, this confirms that the WBF and riming processes are efficient and responsible for the 475 decrease of LWC compared to adiabatic values. These statements are in agreement with the study from (Jackson 476 et al., 2012) who showed for several boundary layer MPCs over Barrow, Alaska during the ISDAC campaign

477 that the subadiabatic profile of LWC and the decreasing droplet concentration at cloud top may be associated to 478

the ice crystal growth processes involving the liquid phase (riming and WBF) and/or the entrainment of dry air

- 479 from above.
- 480

481 However, Figs. 4 and 5 also showed significant differences in cloud vertical profiles from one regime to another. 482 The COLD situations exhibit the largest values for ice properties (IWC up to 0.075 g m<sup>-3</sup>, N<sub>i</sub> up to 8 L<sup>-1</sup>) together 483 with the lowest LWC values ( $< 0.1 \text{ g m}^{-3}$ ). On the contrary, the WARM\_SO profiles are characterized by the 484 largest liquid droplet concentrations, extinction coefficient and LWC values (~200 cm<sup>-3</sup>, 40 km<sup>-1</sup> and 0.3 g m<sup>-3</sup> 485 respectively) and low values of IWC, extinction and size of ice crystals (IWC < 0.01 g m<sup>-3</sup>,  $\sigma_i < 0.2$  km<sup>-1</sup> and D<sub>eff</sub> 486 < 50 µm respectively). Thus in the WARM\_SO regime, it seems that the ice crystal number is too low and their 487 size too small to efficiently consume liquid droplets by WBF or riming processes (Pruppacher and Klett, 488 1978), explaining on one hand the prevalence of the liquid phase and the other hand that the precipitating ice 489 crystal below the cloud do not reach the surface. Moreover, the habit classification as a function of the 490 temperature shows differences between the COLD regime and the WARM regimes (not shown here). This 491 concern in particular the presence of some large droplets in the WARM regimes which are not present in the 492 COLD regime, and the presence of plate and stellar particles below  $-10^{\circ}$ C or around  $-4^{\circ}$ C, which is consistent 493 with the classical ice crystal morphology diagram ((Libbrecht, 2005; Nakaya, 1954).

494 The adiabatic ratio, shown in Figure 7, confirms this assumption where larger values are encountered for the 495 WARM\_SO situations. Indeed, a large adiabatic ratio denotes that processes responsible for the depletion of 496 liquid droplets (mainly riming or WBF) are relatively less efficient.

497 The ice crystal properties relative to the WARM NO situations are similar to the WARM SO cases, except for 498 the effective diameter where values are similar to the COLD regime ( $D_{eff,i} > 100 \mu m$ ). The liquid droplets for this 499 regime exhibit the lowest concentrations (< 100 cm<sup>-3</sup>), and intermediate LWC value (around 0.2 g m<sup>-3</sup>).

500

501 The meteorological classification used in our study is also based on the air mass origin since it shall impact the 502 cloud microphysical properties, as shown in Gultepe and Isaac (2002). In particular COLD and WARM\_NO 503 situations characterized by northern air mass origin should be associated to more pristine conditions and drier air 504 compared to the WARM SO situations. Airborne in situ aerosol measurements were only available during the 505 POLARCAT 2008 campaign (with particle counters). However nearly continuous aerosol measurements (with 506 particle counters and sizers) but ground based were performed at the Mountain Zeppelin station (Ny-Alesund, 507 Svalbard 475 m above sea level, 79°N, 12°E) ) during a period encompassing the ASTAR and SORPIC 508 campaigns. Even though, these measurements do not provide an accurate estimate of the aerosol concentration at 509 the exact location and time when the clouds were sampled, it still give an indication on the background aerosol 510 loading. Based on these measurements, the mean aerosol number concentrations were 230, 120 and 330 cm<sup>-3</sup> for 511

the COLD, WARM NO and WARM SO respectively we can conclude that pristine conditions are encountered 512 for air masses originating from the North, and that cloud measurements performed under WARM\_SO conditions

513 are more likely to be affected by long range transport of pollution for the South/East.

514 The prevalence of the ice phase for the COLD regime is thus consistent both with the cold temperature and the

515 pristine conditions associated with Northern air masses. Despite similar air mass origins, the WARM NO cases

516 exhibit smaller concentration of ice crystals than the COLD situations. This suggests that the influence the cloud

- 517 top temperature prevails to promote the growth or production of ice crystals. The WARM\_SO cases which 518 combine warm temperatures and continental air masses clearly shows that the ice crystal growth or production is 519 reduced, as well as the precipitation efficiency, and that the liquid phase dominates the cloud structure.
- 520 Additionally, the comparison of the vertical profiles of MPC properties of the present work to the previous
- 521 studies concerning the Western Arctic in section 3.2 showed that the cloud properties for the COLD and 522 WARM NO situations agree with that of M-PACE (REF), in particular in terms of ice concentration and IWC.
- WARM\_NO situations agree with that of M-PACE (REF), in particular in terms of ice concentration and IWC.
  The WARM\_SO cases agree more with the ISDAC situations, in particular the low ice concentration. Jackson
- 524 et al. (2012) explained the very low ice concentration observed during ISDAC as a consequence of more
- 525 polluted situations encountered (compared to M-PACE) that might reduce the secondary ice crystal production 526 efficiency (thermodynamic indirect effect). This conclusion is thus in accordance with our assumption that the
- 527 air mass coming from the South may be more impacted by pollution and may reduce the ice growth efficiency.
- These analysis show that microphysical properties of Arctic MPCs over the Greenland and Norwegian seas are closely linked to the cloud top temperature regime and the environmental conditions such as the air mass origin. Similar conclusions have already been made for MPCs in the western Arctic regions by **Gultepe and Isaac** (2002) who demonstrated the impact of the air mass origin (Pacific Ocean or Arctic Ocean) on the MPC microphysical properties.
- However, a more thorough analysis involving collocated *in situ* aerosols measurements are obviously needed to comfort these findings. For instance, our results are somehow consistent with Lance et al. (2011) or Rangno and Hobbs (2001) who showed that "polluted" MPCs exhibit higher droplet concentrations and smaller ice precipitating particles compared to "clean" MPCs. A large number of droplets is expected to reduce the riming process and thus contribute to the large observed values of LWC (as liquid droplets are not consumed by the ice crystals).
- 539
- 540 To go further into the analysis of our microphysical dataset, additional measurements of key parameters are 541 necessary. In particular, quantifying the mechanisms responsible for the formation and growth of droplets and 542 ice crystals within MPCs by measuring the numbers of ice nuclei (IN) and CCN are needed. It would enable us 543 to perform an accurate ice closure and to quantify for example the possible impact of secondary ice production 544 process). A better characterization of the dynamical processes at cloud scale, with accurate high spatial 545 resolution measurements of vertical wind velocities into and around the MPCs would also be necessary. For 546 instance, upward air motion and turbulent entrainment of air from above the cloud are critical to maintain liquid 547 water in MPCs. Accurate humidity measurements would also be needed to better identify condensational growth 548 of ice crystals (WBF process or direct condensation of water vapor on ice, as described by Korolev (2007)) and 549 resolve the issue of turbulence and mixing at cloud edges and into cloud. All these parameters, along with 550 radiative flux measurements, are of primary importance to constrain our assumptions on the microphysical 551 processes.
- At last, coupling our results (and further observations with new parameters and improved instrumentation) with modeling is of course the best way to quantify the relative impact of each process on the MPC lifetime. But such a work remains beyond the scope of the present study.
- 555
- 556 4. Parameterizations of key microphysical parameters

In section 3, we have shown that *in situ* data provide a detailed characterization of the microphysical and optical properties of MPCs. These measurements can also be used to develop cloud parameterizations and to evaluate remote sensing retrieval products or modeling outputs. This section focuses on the key properties and hence parameters which must be better quantified (**Morrison and Pinto, 2006**), namely: (i) IWC (and LWC) – extinction coefficient relationships, (ii) the variability of the ice and liquid water paths, (iii) the temperature dependent ice crystal concentration and (iv) the liquid water fraction (ratio of LWC over total water content) as a function of the cloud level or temperature.

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#### 4.1. Ice and liquid water contents and integrated paths

Linking cloud microphysical and optical properties is an important step in order to model the cloud radiative properties or to constrain/develop remote sensing retrieval methods. In particular, accurate IWC-extinction relationships and integrated properties such as ice and liquid water paths are needed to improve the remote sensing retrieval products and cloud modeling (**Heymsfield et al., 2005; Waliser et al., 2009**). In this section, we provide such relationships and parameters based on *in situ* measurements.

573

Figures 8a and 8b display the IWC and the LWC measurements as a function of the ice and droplet extinction coefficients respectively, with the temperature superimposed in color. The averaged values of IWC and LWC over intervals of 0.1 km<sup>-1</sup> and 2 km<sup>-1</sup> for the ice and liquid extinction coefficient respectively are represented by the grey squares in order to determine the fitting curves (represented by the red lines with the mean absolute error in dashed lines, see Eqs. (3) and (4) below). Ice crystals and liquid droplets extinction coefficients are well correlated with their water content counterparts. The correlation coefficients are high (0.88 for ice and 0.89 for liquid) and the IWC-σ relationships are nearly linear.

It should also be noted that including the temperature as an additional parameter for the linear fitting did not improve the accuracy of the parameterizations, contrary to previous studies of **Heymsfield et al. (2005)**, **Hogan et al. (2006)**, or **Protat et al. (2007, 2016)**. However these previous studies concerned tropical and mid-latitude clouds and cover a much broader range of temperatures (from -65° °C to 0 °C, compared to narrower range from 24°C to 0 °C in our study).

- 587  $IWC = 0.076 \sigma^{1.06}$  (3) 588  $LWC = 0.0016 \sigma^{1.31}$  (4)
- $589 \qquad \text{with IWC and LWC in g m}^{-3} \text{ and } \sigma \text{ in km}^{-1}.$
- 590

586

591 Integrated properties such as LWP and IWP are common modeling outputs which suffer from large 592 discrepancies depending on the model specifications (**Waliser et al., 2009**). Moreover only a very limited 593 number of studies were devoted to retrieve these properties from *in situ* measurements in this region of the 594 Arctic. Since the flight legs selected in our study target ascending and descending sequences into single-layer

- 595 MPCs, *in situ* measurements can be used to determine IWP and LWP according to the following equation:
- 596

597 IWP (or LWP) = 
$$\int_{ground}^{cloud\ top} IWC\ (or\ LWC)(z)dz$$

599 We recall that these integrated properties should be considered as quasi-instantaneous as ascending and 600 descending flight sequences are obviously not fully vertical and need about 5-10 minutes to be performed 601 (compared to the snapshots performed by remote sensing measurements).

(5)

602

603 Figure 8c displays the ice (green) and liquid (blue) water paths as a function of the cloud top temperature (1 °C 604 intervals). For cloud top temperatures below -20 °C, IWP and LWP reach values close to 30 g m<sup>-2</sup> and 50 g m<sup>-2</sup> 605 respectively. The IWP decreases dramatically when the cloud top temperature increases: to very low values close 606 to 0 are encountered at temperatures above -8 °C. LWP reaches a maximum of 100 g m<sup>-2</sup> at -13 °C and the 607 smallest values (around 15 g m<sup>-2</sup>) are encountered when the cloud top temperature is typically around -18°C. 608 These findings are consistent with the main previous studies devoted to Arctic MPCs from (Hobbs et al., 2001; 609 Pinto, 1998; Pinto and Curry, 2001; Shupe et al., 2006). They reported mean LWP values in the range of 20-610 70 g m<sup>-2</sup>, with some maxima up to around 130 g m<sup>-2</sup>, and IWP mean values less than 40 g m<sup>-2</sup>. However, one 611 shall note that all these previous studies concerned once again the MPCs in the western Arctic regions (Barrow, 612 Alaska, Beaufort Sea).

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### 4.2. Ice crystal concentration

616 The accurate knowledge of the ice crystal concentration is of primary importance to correctly parameterize the 617 initiation and evolution of the ice phase in models, and reduce the significant uncertainties in the modelling of 618 the ice/liquid partitioning within MPCs.

619 Figure 9 shows the maximum number concentration of ice crystals with size greater than 100 µm as a function of 620 the cloud top temperature for each MPC vertical profile. The data points are color-coded according the COLD, 621 WARM\_NO and WARM\_SO environmental regimes. This figure highlights that the maximum ice 622 concentration varies almost exponentially (figure is in log-lin scale) with the cloud top temperature, with 623 nevertheless a large variability. Thus, a relationship may be fitted in order to parameterize the ice crystals 624 number concentration as a function of temperature in MPCs (Eq. (6), also included in Fig. 9), even though the 625 correlation coefficient is quite low (0.43). The mean absolute error (MAE) is also displayed on figure 9 (dotted 626 lines) to estimate the uncertainties on the parameterization.

627

628 
$$N_{i,max} = e^{(-0.191 T_{top} - 1.134)}$$
 with N<sub>i,max</sub> in L<sup>-1</sup> and T<sub>top</sub> in °C (6)

629

630 For comparison purposes, the parameterizations of Meyers et al. (1992) and Cooper (1986) for heterogeneous 631 ice nucleation and the parameterization of Young et al. (2017) for primary ice nucleation based on 632 microphysical observations during the Aerosol-Cloud Coupling and Climate Interactions in the Arctic 633 (ACCACIA) campaign are displayed (in purple, orange and brown dashed lines respectively). The Meyers et al. 634 (1992) parameterization is within the range of the uncertainties of our parameterization. However it significantly 635 deviates from our relationship for cloud top temperature higher than -15°C i.e. for clouds under warm regime. 636 For these regimes the ice number concentrations can differ by a factor up to two at  $-10^{\circ}$ C.

The parameterizations of **Cooper (1986)** and **Young et al. (2017)** do not match with the present parameterization since the ice crystal concentrations predicted are around one order of magnitude lower than the ones in the present study. This difference can be explained by the different seasons, cloud types and locations of the observations used for the parameterization of **Cooper (1986)** and the fact that the range of their measured concentrations lie within a factor of 10 as they noted.

642 In contrast, the sampling conditions for the determination of the Young et al. (2017) parameterization are more 643 similar to the present work, they used measurements in Arctic MPCs over the Greenland Sea. The dataset was 644 collected during spring and summer, above open sea, ice sheet and transitions. This variability in the seasons and 645 surface conditions may explain the differences observed compared to the present work. Above all, Young et al. 646 (2017) displayed an averaged concentration whereas the maximum ice number is presented here. But even by 647 taking the averaged ice concentrations in the present work, the parameterization does not match with that of 648 Young et al. (2017) (not shown here). Finally, the detailed time series displayed in the Young et al. (2016) and 649 Lloyd et al. (2015) works which present the cases used for the determination of the parameterization of Young 650 et al. (2017) showed that the maximum ice number concentrations frequently displayed values between 1  $L^{-1}$  and 651 5 L<sup>-1</sup> which is in the range of the present parameterization.

652 Our results could not be compared to more sophisticated parameterizations accounting for supersaturation and 653 aerosol properties (such as (**DeMott et al., 2011**) since additional data are needed (aerosol and CCN/IN 654 measurements, humidity). These additional data are also necessary to discuss the processes such as the secondary 655 ice production processes which could explain the higher crystal numbers observed in the present study compared 656 to the other works presented in this section.

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#### 4.3. Liquid water fraction

659 660

661 The MPC liquid fraction can be determined based on the separate liquid and ice properties presented in section
662 3. The liquid water fraction (hereafter LWF) is defined as the ratio of liquid water content LWC over the total
663 water content TWC (IWC+LWC) at each altitude level.

To our knowledge very few previous studies have assessed the liquid water fraction in MPCs. Most of them
concerned MPCs only in western Arctic regions (de Boer et al., 2009; McFarquhar et al., 2007; Shupe et al.,
2006).

Figure 10a displays the liquid fraction according to the normalized altitude  $Z_n$ . For purpose of comparisons, the parameterization from **McFarquhar et al. (2007)** (hereafter MF07) determined from *in situ* measurements during M-PACE is also represented on Fig. 10a by the black dotted curve. Our relationship (Eq. (7)) significantly deviates from that of MF07.

671

672  $LWF = 28.4 Z_n + 54.9$  with LWF in % (7)

673

They used *in situ* measurements from 53 profiles in single layer MPCs sampled over Alaska with temperatures
ranging from -3 °C to -17 °C. As mentioned in section 3 they observed similar ice crystal number concentrations
but ice crystals smaller with mean effective diameters around 50 μm compared to 100 μm in our study.

Figure 10b shows the liquid fraction according to cloud top temperature. Each point represents the mean value of the liquid fraction determined for each profile. The error bars corresponding to the standard deviation display large values around 80 % indicative of a large variability. Nevertheless, Fig. 10b shows that LWF is well correlated with the cloud top temperature (Eq. (8)). The decrease in LWF associated with a decrease of temperature is consistent with Fig. 9 which shows that ice number concentration increases for colder temperatures.

683

684 
$$LWF = 2.97 T_{top} + 121.20$$
 with LWF in % and  $T_{top}$  in °C (8)

685

690

686 The liquid fraction is also determined at each cloud level as a function of the temperature on Fig. 10c (with 1 °C 687 temperature interval). The same trend as in Fig. 10a is observed. The liquid water fraction increases with 688 decreasing temperature. The relationship between LWF and T is nearly linear with similar slopes for the three 689 regimes (Eqs. (9a), (9b) and (9c) for the COLD, WARM\_NO and WARM\_SO regimes respectively).

- 691LWF = -3.02T + 3.95(9a)692LWF = -3.48T + 47.60(9b)693LWF = -1.70T + 76.16(9c)
- $694 \qquad \text{with LWF in \% and T in }^\circ\text{C}.$
- 695

However, large shifts are observed from one regime to another, especially when comparing the COLD regime to
the WARM\_NO and the WARM\_SO. This shift is clearly linked to the temperature profiles (see Fig. 3).
However, one can note that the results for the WARM\_NO regime are the ones in the closest agreement with the
MF07 parameterization.

In order to compare our results to those of **Shupe et al. (2006**), we also determined the total liquid water fraction (LWF<sub>total</sub>) in terms of water paths (LWP/TWP). Fig. 10d shows a rather good agreement between the two water path ratios, showing that IWP dominates in the coldest clouds ( $T_{top}$  around -20 °C in average). On the opposite, LWF<sub>total</sub> is more important in the warmer MPCs ( $T_{top}$  above -15 °C). However such liquid fraction determination must be taken with care since it integrates the ice region below the clouds (**de Boer et al., 2009**).

705 706

# 707 5. Conclusions and outlook

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In this study, a characterization of Arctic boundary-layer mixed-phase clouds microphysical properties has been performed. *In situ* data from 4 airborne campaigns over the Greenland Sea and the Svalbard region are compiled and analyzed. The data set represents in total 18 flights and 71 vertical profiles in MPCs (more than 350 minutes of cloud *in situ* observations). Cloud phase discrimination is achieved and vertical profiles of number, size, mass and shapes of ice crystals and liquid droplets within MPCs are determined.

714

715 The main conclusions of the present work are summarized as follow:

717i)Liquid phase is mainly present in the upper part of the MPCs with high concentration of small718droplets ( $N_w \sim 120 \text{ cm}^{-3}$ ,  $D_{eff,w} \sim 15 \mu m$ ), and averaged LWC around 0.2 g m<sup>-3</sup>. Ice crystals are719present everywhere in the MPCs with no significant vertical variability ( $N_i \sim 3 \text{ L}^{-1}$ ,  $D_{eff,i} \sim 100$ 720 $\mu m$ , IWC ~ 0.025 g m<sup>-3</sup>), but mainly in the lower part, and precipitate down to the surface. The721morphology study of ice crystals images showed that irregular and rimed particles prevail over722stellars and plates habits.

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- 724 The vertical profiles of the microphysical properties and the shape distribution can also be ii) 725 used to give an insight of the microphysical processes occurring in MPCs. It is likely that 726 adiabatic lifting (condensation) is the main process for liquid droplets initiation and growth, 727 and that evaporation at cloud top due to entrainment of dry air seems to occur. In the cloud 728 layer, where liquid droplets and ice crystals coexist, Wegener-Bergeron-Findeisen and riming 729 processes are the main mechanisms involved in the ice crystal growth. The large occurrence of 730 irregular particles highlights the fact that the ice crystals undergo a variety of growth 731 processes, and the turbulence in the MPCs life cycle is efficient to mix the cloud.
- 733 iii) The analysis of the scattering phase function showed a very high correlation between optical734 properties and liquid to ice fraction within the MPC layers.
- 736 iv) Statistical analysis exhibits significant differences in the vertical profiles of MPC properties 737 depending if the cloud top is cold or warm and if the air mass has its origin from higher or 738 lower latitudes. The largest droplet concentration and LWC values observed (200 cm<sup>-3</sup>, 0.3 g 739 m<sup>-3</sup> respectively) are associated with the warm temperature regime with air mass originating 740 from the South/East (continental areas). For these situations, at the same time, very low values 741 of ice crystal size and IWC are observed (IWC < 0.01 g m<sup>-3</sup>,  $D_{eff,i} \sim 50 \mu m$ ). On the opposite, 742 the colder situations exhibit large values of ice contents especially when air masses originate 743 from the North (IWC ~ 0.075 g m<sup>-3</sup>). These results underline the importance of the air mass 744 origin and the cloud top temperature as well as the need of simultaneous aerosol measurements 745 (sources, transport, physical and chemical properties) in connection with the MPC properties 746 to study the cloud-aerosol interactions and improve the understanding of ice and liquid 747 formation processes.
- 749 v) The main results of the present work were compared to the previous studies which concern 750 mainly MPCs in the Western Arctic region. The main findings showed that the properties of 751 the COLD and WARM NO situations (large values of ice properties) of the present work are 752 consistent with the rather clean situations of previous Western Arctic studies such as M-PACE. 753 On the opposite, the MPC properties of the WARM SO cases (prevalence of liquid phase and 754 very low values of ice properties) are more in agreement with the more polluted situations in 755 Western Arctic, such as ISDAC. These findings confirm that the MPC properties are strongly 756 linked to the environmental conditions such as temperature and air mass origin.

- vi) Several parameterizations for remote sensing or modeling are proposed. It concerns the determination of IWC (and LWC) extinction relationships, ice and liquid integrated water paths, the ice concentration and liquid water fraction. Comparisons with the few previous works available in the literature showed in general a good agreement. Obviously, the application range of the established relationships is only for Arctic MPCs and temperature range between 0 and -23 °C. A next step to the present work will be to apply the proposed parameterizations to remote sensing algorithms and modelling to investigate their relevance.
- 765 766

This study provided for the first time a statistical analysis of Arctic MPC *in situ* data from 4 airborne campaigns
located in the Eastern Arctic region. An accurate characterization of the vertical variability of liquid droplets and
ice crystals properties has been made, allowing the development of parameterizations.

770 Further studies should involve new measurement technic to provide accurate characterization of cloud phase and 771 microphysical properties, in particular for the small particles. This will allow to complete and validate the 772 present results. For example, instruments like the small ice detector (SID-3, Ulanowski et al., 2014; Vochezer 773 et al., 2016) or the cloud particle spectrometer with polarization detection (CPSPD, Baumgardner et al. (2014)) 774 should provide valuable measurements to differentiate droplets from ice crystal even at size lower than 50µm. 775 Both probes are open path to avoid shattering artefacts. Additionally, accurate measurements of humidity and 776 aerosol (CCN and IN) remains an important shortage in order to deep the analysis of microphysical processes 777 and to realize ice and liquid closure and better understand life cycle and persistence of such particular clouds. In 778 this purpose, a modeling study of cloud microphysics shall be of help. Finally, by characterizing clouds at very 779 low altitude levels, this work can be useful in future studies for validate/evaluate space remote sensing 780 observations and retrieval products (A-Train, EarthCare...) since these measurements are known to have 781 important shortcomings near the surface.

- 782
- 783

# 784 Appendix A: Data processing of *in situ* measurements

785

The methodology developed by Lawson and Baker (2006) to derive the Ice Water Content (IWC) from 2D
particle images recorded by the CPI instruments is applied (Eq. (A1) below).

788 
$$IWC = \frac{0.135 \sum_{i} X_{i}^{0.793}}{V}$$
 (A1)

789 where *V* is the sample volume and  $X_i$  is the mass parameter for each crystal image defined by Lawson and Baker 790 (2006) as follow:

791 
$$X_i = \frac{A_i \times W_i \times 2 \times (L_i + W_i)}{P_i}$$
(A2)

A<sub>i</sub>,  $W_i$ ,  $L_i$  and  $P_i$  are the area, width, length and perimeter of the crystal image *i*.

The extinction coefficient ( $\sigma$ ) and the effective diameter (D<sub>eff</sub>) are determined from CPI and FSSP measurements as follow:

796 
$$\sigma_{ice\ (or\ liquid)} = \mathbf{2} \times \frac{\sum_{i} A_{i}}{v}$$
 (A3)

797 
$$\boldsymbol{D}_{eff,ice\ (or\ liquid)} = \boldsymbol{C} \times \frac{IWC\ (or\ LWC)}{\sigma_{ice}\ (or\ \sigma_{liquid})}$$
(A4)

798 with constant  $C = 3000 \text{ mm}^3 \text{ g}^{-1}$  according to **Gayet et al. (2002)**.

The LWC derived from the Nevzorov probe measurements is calculated according to Korolev et al. (1998) :

800

801 
$$LWC_{Nevzorov} = \frac{P_{LWC} - (\frac{P_{TWC} \times \varepsilon_{LWC,i} \times S_{LWC}}{\varepsilon_{TWC,i} \times S_{TWC}})}{L_{v} \times S_{LWC} \times U \times (\varepsilon_{LWC,i} - \frac{\varepsilon_{LWC,i} \times \varepsilon_{TWC,i}}{\varepsilon_{TWC,i}})}$$
(A5)

802

803 where  $P_{LWC}$  and  $P_{TWC}$  are the power supplied to the LWC and TWC sensors to maintain the constant temperature 804 of the wire.

 $S_{LWC}$  and  $S_{TWC}$  are the surface of the sensors,  $L_v$  is the latent heat of vaporization and U is the true airspeed.

806 The epsilon terms refer to the collection efficiencies of liquid droplets (l index) or ice crystals (i index) on the

807 LWC and TWC sensors. These efficiencies are set as follow:

808  $\epsilon_{LWC,1} = 0.76$  : see Schwarzenboeck et al. (2009);

809  $\epsilon_{LWC,i} = 0.11$  : following Korolev et al. (1998);

810  $\varepsilon_{TWC,1} = 1$ : according to **Korolev et al. (1998)** for droplets with size around 25 µm;

811  $\epsilon_{TWC,i} = 1$ : following Schwarzenboeck et al. (2009). It should be noticed that taking  $\epsilon_{TWC,i} = 3$  (as

812 assumed in **Korolev et al., 2013**) instead of 1 induces an increase of LWC by 10 % only.

813 The uncertainties associated to the microphysical and optical properties derived from FSSP-100, PN, Nevzorov

814 and CPI measurements are detailed in (Baumgardner and Spowart, 1990; Gayet et al., 2002; Korolev et al.,

815 **1998; Mioche, 2010**) respectively, and are summarized in Table 2.

# 816 Appendix B: Effects of shattering of ice crystals on measurements

Techniques and methods exist now to avoid or estimate this shattering effect, such as new-designed inlets or measurements of the particles inter-arrival time (**Field et al., 2003**), but none of these were available for this study. However in order to assess the accuracy of the present dataset and highlight a possible impact of shattering effect, a brief intercomparison of the extinction coefficient from the three data sets was conducted. Indeed, the extinction coefficient is the only parameter which can be derived by the measurements of the three probes. Moreover, it is not determined with the same method, since it is calculated from the PSD for the CPI and the FSSP, and from the scattering phase function for the PN. One more important point is that CPI, FSSP and PN

- have all different size inlets (23 mm, 40 mm and 10 mm diameter respectively). So, from these information, we
- 825 could assume that, if shattering effect is present on ice particles, its magnitude (i.e. the number of smaller new 826 artifact particles) would differ from one instrument to another. Thus, the comparison of the extinction coefficient
- artifact particles) would driver from one instrument to another. Thus, the comparison of the extinction events
- 827 from CPI, FSSP and PN measurements would highlight such discrepancies.
- 828 Figure B1 displays the comparison of the extinction coefficient derived from the PN and from the combination
- 829 of the CPI and FSSP for all the *in situ* data available for this study. Note that the combination of CPI and FSSP
- 830 data covers the same size range of the PN. Figure B1 clearly shows that the extinction coefficient measurements
- 831 derived from the combination of the CPI and FSSP and the PN are very well correlated (with a coefficient of
- 832 0.87) and no significant bias is observed (regression coefficient of 0.98). Thus, since the design of the
- 833 instruments and data processing are different for each dataset, these results highlight that the shattering effect is
- probably smaller than the measurements uncertainties (25 %, 35 % and 55 % for PN, FSSP and CPI respectively,
- see Table 2).

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1186 Table 1: Summary of *in situ* observations of Arctic single layer MPCs.

Field experiment	Location [latitude range]	Date	Number of flights in MPCs	Number of profiles in MPCs	Duration of data (minutes)
ASTAR 2004	Spitzbergen (Norway) [76-79]° N	May 2004	4	7	30
ASTAR 2007	Spitzbergen (Norway) [76-79]° N	April 2007	5	34	173
POLARCAT 2008	Kiruna (Sweden) [68-73]° N	April 2008	4	10	45
SORPIC 2010	Spitzbergen (Norway) [75-78]° N	May 010	5	20	109
TOTAL	T	I	18	71	357

1190 Table 2: Uncertainties of cloud properties derived from CPI, FSSP, PN and Nevzorov instruments.

Probe [Measurements range]	Number concentration (N)	Extinction coefficient (σ)	Effective diameter (D <sub>eff</sub> )	Water contents (IWC or LWC)	Asymmetry parameter (g)
CPI [15 µm to 2.3 mm]	50 %	55 %	80 %	60 %	-
FSSP-100 [3 to 45 µm]	10 %	35 %	4 %	20 %	-
PN [< 800 μm]	-	25 %	-	-	4 %
Nevzorov [LWC > 0.003-0.005 g m <sup>-3</sup> ]	-	-	-	20%	-

1195 Table 3: Statistics of cloud base and cloud top altitudes along with cloud layer thickness obtained from the 71 profiles

sampled in MPCs.

	Mean	Standard dev.	Median	25 <sup>th</sup> percentile	75 <sup>th</sup> percentile	Max.	Min.
z <sub>top</sub> (m)	1200	310	1200	1000	1370	2120	525
z <sub>base</sub> (m)	756	283	700	510	850	1700	400
Layer thickness (m)	444	211	420	270	600	950	100



Table 4: Summary of the method for the assessment of the cloud thermodynamical phase and liquid droplet and ice crystal properties from the combination of PN, CPI, FSSP and Nevzorov probes.

	[c	PN g-values orresponding cloud phas	se]
	g < 0,80 [ice]	0,80 < g < 0,83 [mixed]	g > 0,83 [liquid]
Instrument [measurement range]			
FSSP [15 to 45 μm]	NO	YES	YES
Nevzorov probe [LWC > 0.003-0.005 g m <sup>-3</sup> ]	NO	YES	YES
CPI [15 µm to 2.3 mm]	YES	YES	NO

Experiment	Date	Mean T <sub>Top</sub> (°C)	Air mass origin N = North (Arctic Ocean) S/E = South or East (Continental	Regime
ASTAR 2004	15 may	-16,5	Ν	COLD
	22 may	-8,5	S	WARM_SO
	25 may	-8	Ν	WARM_NO
	5 june	-11	Ν	WARM_NO
ASTAR	2 april	-21	Ν	COLD
	3 april	-16	Ν	COLD
	7 april	-22	Ν	COLD

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1<sup>srt</sup> april

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COLD

COLD

WARM\_NO

WARM\_NO

WARM\_NO

WARM\_NO

WARM\_SO

WARM\_SO

WARM\_SO

WARM\_SO

WARM\_NO

Table 5: Classification of the MPC situations according to temperature regimes and air mass origins

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1214 Figure 1: Location of the MPC measurements during the ASTAR, POLARCAT and SORPIC campaigns.





1218Figure 2: a): Normalized scattering phase function according to the normalized altitude from Polar Nephelometer1219measurements (few μm to around 800 μm size range), averaged over all the campaigns. g-values indicate the cloud1220phase: g < 0.80: ice, 0.80 < g < 0.83: mixed and g > 0.83: liquid. b): Mean vertical profile of asymmetry parameter (for1221all the campaigns). The grey bars indicate the threshold g-values for the assessment of ice, mixed and liquid cloud1222phases.



1226Figure 3: Vertical profiles (normalized altitude) of the mean temperature for each regime. Shaded spreads represent1227the standard deviation. The mean cloud base and top altitudes and their standard deviation for each regime are1228indicated.



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Figure 4: Vertical profiles (expressed in normalized altitude) of liquid droplets properties from FSSP or Nevzrov probe measurements (3-45 μm size range): a) extinction coefficient, b) droplet concentration, c) LWC, d) effective diameter for the three regimes and averaged over all the campaigns and e) averaged droplet size distribution for all the campaigns.

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1241 Figure 5: Vertical profiles (expressed in normalized altitudes) of ice crystal properties from CPI measurements (15

μm - 2.3 mm size range): a) extinction coefficient, b) ice crystal concentration, c) IWC, d) effective diameter for the
 three regimes and averaged over all the campaigns and e) averaged particle size distribution for all the campaigns.



Figure 6: Vertical profiles of particle shapes (from CPI measurements and for particles larger than 100 μm)
 according to normalized altitude (top panels) and temperature (bottom panels). Distributions are displayed according
 to particle number (left panels) and mass (right panels).





1256 Figure 7: Vertical profiles of the ratio of measured LWC over theoretical adiabatic LWC for the three regimes.



- 12: -
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Figure 8: a) IWC and b) LWC as a function of extinction coefficient. Color scale is the temperature, grey squares represent the values averaged over extinction coefficient intervals of 0.1 km<sup>-1</sup> and 2 km<sup>-1</sup> for IWC and LWC respectively. The red lines represent the curve fittings and the dashed-line the uncertainties on the fitted relationships (Mean Absolute Errors). c) Ice (green) and liquid (blue) water paths according to the cloud top temperature.

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1268Figure 9: Maximum ice crystal concentration as a function of cloud top temperature. The colored circles represent the1269values for each profile (with fitting in black solid line and mean absolute error in dotted lines). The Meyers et al.1270(1992), Cooper (1986) and Young et al. (2017) parameterizations are also displayed in purple, orange and brown1271dashed lines respectively.



1275Figure 10: Liquid water fraction according to  $Z_n$  (a), cloud top temperature (b) and temperature (c). The dotted1276dashed line on panels a) and c) is the parameterization from McFarquhar et al. (2007) and the solid lines on panels a),1277b) and c) are the fittings for the present study. d) Ratio of LWP over TWP according to cloud top temperature. The1278solid line refers to the present study and the dotted lines refers to Shupe et al. (2006) work.



1283Figure B1: Comparison of extinction from PN and CPI+FSSP measurements. Grey bars represent the 25 %1284uncertainties on the PN extinction. The red dotted line is the linear fitting (slope of 0.98,  $R^2 = 0.87$ ) and the blue dotted1205

1285 line is the 1:1 line.