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Characterization of the cloud microphysical and optical properties and aerosol-cloud interaction in the Arctic from in situ ground-based measurements during the CLIMSLIP-NyA campaign, Svalbard

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1 **Characterization of the cloud microphysical and optical**
2 **properties and aerosol-cloud interaction in the Arctic from**
3 **in situ ground-based measurements during the CLIMSLIP-**
4 **NyA campaign, Svalbard**

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6
7 Gwennolé GUYOT¹, Frans OLOFSON¹, Peter TUNVED², Christophe GOURBEYRE¹, Guy
8 FEVBRE¹, Régis DUPUY¹, Christophe BERNARD¹, Gérard ANCELLET³, Kathy LAW³,
9 Boris QUENNEHEN⁴, Alfons SCHWARZENBOECK¹, Kostas ELEFThERIADIS⁵, Olivier
10 JOURDAN¹

11
12 ¹ Laboratoire de Météorologie Physique (LaMP), Université Blaise Pascal (UBP), OPGC,
13 CNRS UMR 6016, Clermont-Ferrand, France

14 ² Department of Applied Environmental Science (ITM), Stockholm University, Stockholm,
15 Sweden

16 ³ Laboratoire Atmosphère, Milieux et Observations Spatiales (LATMOS), IPSL, UPMC,
17 CNRS UMR 8190, Paris

18 ⁴ Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), Université
19 Grenoble Alpes/CNRS, 38041 Grenoble, France

20 ⁵ Environmental Radioactivity Laboratory, Institute of Nuclear and Radiological Science &
21 Technology, Energy & Safety, Attiki, Greece

22
23 **Abstract**

24
25 This study will focus on cloud microphysical and optical characterization of three different
26 types of episodes encountered during the ground based CLIMSLIP-NyA campaign performed
27 in Ny-Alesund, Svalbard: the Mixed Phase Cloud (MPC), snow precipitation and Blowing
28 Snow (BS) events. These in situ cloud measurements will be combined with aerosol
29 measurements and air mass backtrajectory simulations to qualify and parameterize the arctic
30 aerosol cloud interaction and to assess the influence of anthropogenic pollution transported into
31 the Arctic.

32 The results show a cloud bimodal distribution with the droplet mode at 10 μm and the crystal
33 mode centered at 250 μm , for the MPC cases. The precipitation cases presents a crystal
34 distribution centered around 350 μm with mostly of dendritic shape. The BS cases show a
35 higher concentration but smaller crystals, centered between 150 and 200 μm , with mainly
36 irregular crystals.

37 A “polluted” case, where aerosol properties are influenced by anthropogenic emission from
38 Europe and East Asia, was compared to a “clean” case with local aerosol sources. These
39 anthropogenic emissions seem to cause higher Black Carbon, aerosol and droplet
40 concentrations, a more pronounced accumulation mode, smaller droplet sizes and a higher
41 activation fraction F_a . Moreover, the activation diameter decreases as the droplet diameter
42 increases and F_a increases showing that smaller particles are activated and droplets grow when
43 the aerosol number decreases. This is in agreement with the first (Twomey) and second
44 (Albrecht) aerosol indirect effect. The quantification of the variations of droplet concentration
45 and size leads to IE (Indirect Effect) and NE (Nucleation Efficiency) coefficients values around
46 0.2 and 0.43, respectively. These values are close to those found by other studies in the arctic



47 region which confirms these parameterizations of arctic aerosol-cloud interaction in climate
48 models.

49 **1 Introduction**

50

51 The Arctic is a region where the surface warming is faster than the global average warming,
52 associated with, in particular, a rapid melting of the sea ice in summer (Vaughan et al., 2013).
53 This is the so-called arctic amplification. Several studies indicate that the arctic warming is
54 mainly of anthropogenic origin (e.g., Mc Guire et al., 2006, Serreze and Francis, 2006). The
55 arctic amplification is due to several positive feedbacks specific to the Arctic, the most
56 important being the sea ice melting feedbacks (Screen and Simmonds, 2010). Changes in
57 atmospheric and oceanic circulation, cloud properties (especially cloud cover) and atmospheric
58 water vapor amount are highly expected but their quantification remains uncertain. Specially,
59 the effects of clouds dominate the intermodal standard deviation of a temperature rise due to an
60 increase of atmospheric CO₂ concentration (Dufresne and Bony, 2008).

61

62 Recent remote sensing studies have shown that the clean and stable arctic atmosphere is
63 characterized by a high occurrence of mixed phase clouds (MPC) all year long, except in winter
64 and early spring when ice clouds are important (Mioche et al., 2015). However, the Svalbard
65 region is an exception where MPC are the most frequent cloud independent of season (Mioche
66 et al., 2015). Moreover, the altitude of the MPC is highly dependent of the height of the
67 inversion layer. The frequently occurring situation with a stable atmosphere and the low level
68 pronounced inversion layer promotes low level clouds of stratus form (Curry et al., 1996). The
69 arctic MPC are composed of a liquid layer on top and below which is located the mixed phase
70 where ice crystals take form (Gayet et al., 2009). If the crystals grow enough, a precipitation
71 layer is produced below the cloud. The dynamics together with possible surface coupling and
72 advection are essential to maintain the MPC during several days (Morrison et al., 2012). This
73 structure results from a complex network of interactions between numerous local and larger
74 scale processes that complicates the understanding of the MPC properties evolution and its
75 impact on arctic climate (Morrison et al., 2012). In the Arctic, the umbrella effect is not
76 necessarily dominant compared to the cloud greenhouse effect (Quinn et al., 2008), which
77 suspects that clouds play an important role in the arctic amplification. Several studies have
78 revealed that MPCs have a large impact on the surface radiative flux in the Arctic (e.g. Kay et
79 al., 2012, Wendisch et al., 2013).

80

81 Arctic cloud properties are linked to aerosol properties since they can act as Cloud
82 Condensation Nuclei (CCN) or Ice Nuclei (IN). Thus, aerosol seasonal variability and transport
83 from lower latitudes play a role in cloud properties evolution. Studies have shown an arctic
84 annual mean aerosol concentration half that for mid-latitudes. The stable atmosphere and the
85 dark winter promote growth by coagulation/coalescence of the particles, i.e. an increase in size
86 and decrease in concentration, with dominant accumulation mode (Tunved et al., 2013). When
87 the sun rises during spring, these big particles, which can come from lower latitudes, generate
88 the arctic haze phenomenon (Quinn et al. 2007). The stronger sun light gives rise to increasing
89 photochemical activity associated with new particle formation and a dominant Aitken mode
90 (Engwall et al., 2008). Moreover, the ice melting exposes land surfaces that can act as aerosol
91 sources. Therefore, the aerosol concentration increases until its maximum in summer. These
92 features were observed in Alaska (Quinn et al., 2002) and Svalbard (Tunved et al., 2013),
93 proving that they are representatives of the aerosol properties evolution in the Arctic.

94



95 The rapid change in aerosol properties occurring in spring is known to cause changes in arctic
96 cloud properties, the so-called aerosol indirect effect. Increase in aerosol concentration with
97 constant Liquid Water Path (*LWP*) is known to increase cloud droplet concentration and cloud
98 optical thickness but decrease droplet size (Twomey, 1974, 1977), decrease the precipitation
99 efficiency and increase the cloud lifetime (Albrecht, 1989). Also, in a temperature rise scenario,
100 the cloud height is expected to increase (Pincus and Baker, 1994). The impacts of anthropogenic
101 aerosol transported to the Arctic on clouds are not fully understood, but Garrett and Zhao (2006)
102 showed that the cloud emissivity is higher for polluted case, contributing to the arctic warming.
103

104 In the case of arctic MPC where liquid and ice phases coexist, the aerosol-cloud interaction is
105 complexified by the addition of the ice phase and several interaction mechanisms have been
106 assumed. Lohmann (2002a, 2002b) proposed that an increase in ice nuclei could increase the
107 cloud ice content at the expense of the liquid content. This so-called glaciation indirect effect
108 would mean, as the precipitation is more efficient for the ice phase, a decrease in cloud cover
109 in lifetime. The riming indirect effect predicts a riming efficiency decrease due to the
110 supercooled droplet size decrease. Thus, an increase in Cloud Condensation Nuclei (CCN)
111 could lead to a decrease in Ice Water Content (IWC) and ice particles concentration (Borys et
112 al., 2003). According to the data of the two measurement campaigns ISDAC (Indirect and Semi-
113 Direct Aerosol Campaign) and MPACE (Mixed-Phase Arctic Cloud Experiments), Jackson et al.
114 (2012) found a correlation corresponding to the glaciation effect above the cloud liquid phase
115 but no evidence of the riming effect. Mc Farquhar et al. (2011) showed that the aerosol size is
116 the main parameter to explain the particles activation and that the chemical properties don't
117 determine the ability of an aerosol to act as a CCN, i.e. the Kelvin effect is dominant compared
118 to the Raoult effect in the Arctic.
119

120 The work presented here is included in the frame of the project CLIMSLIP (CLimate IMpacts
121 of Short-Lived Pollutants in the polar region). The main objective of this project is to reduce the
122 uncertainties of the radiative forcing due to the anthropogenic emissions of tropospheric ozone,
123 methane and aerosol including Black Carbon (BC). This article will focus on the arctic ground
124 based in situ cloud and aerosol measurement study, performed at the Mount Zeppelin station
125 (474 meters altitude), in Ny-Alesund, Svalbard, performed during spring 2012. First, a
126 classification and characterization of the different types of cases will be presented. Then, a
127 comparison between a polluted and a clean case will be made, based on air masses
128 backtrajectories. In the end, the different aerosol-cloud interactions will be discussed and, if
129 possible, quantified.
130

131 **2 Site & instrumentation**

132

133 **2.1 Site**

134

135 The campaign was carried out between February 26 and May 2 at the Mount Zeppelin station
136 (78°56'N, 11°53'E) located south-west of the Ny-Alesund village, Svalbard, at an altitude of 474
137 meters above sea level. This station presented in Figure 1 was built and is managed by the
138 Norwegian Institute for Air Research (NILU). The Zeppelin observatory is mostly unaffected by
139 local sources and is considered to be within the boundary layer most of the time (Tunved et al.,
140 2013). This station represents remote arctic conditions and is a part of the European observation
141 network ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure network).
142 Continuous measurements of atmospheric trace gases and aerosol physical and chemical properties



143 are performed all year long. The station is also equipped with instruments to measure temperature,
144 humidity and wind speed and direction.
145 A ceilometer, CL51 model, was installed in the Ny-Alesund village at sea level. This remote
146 sensing instrument is designed to measure the clouds within an altitude range between 0 and 15
147 km. It uses the technology of a lidar with a laser wavelength at 910 nm. During CLIMSLIP, the
148 ceilometer was used to retrieve the approximate altitude of the mixed phase and the liquid layers
149 and showed good agreement with the microphysical measurements. However, in some cases,
150 fog or an optically thick ice layer prevents the laser beam from penetrating within the cloud
151 system.

152
153



Figure 1: Picture of the Mount Zeppelin station (www.npolar.no)

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157 2.2 Instrumentation and data processing

158

159 2.2.1 Cloud instrumentation

160

161 The cloud ground based instrumentation used during CLIMSLIP-NyA was installed on a
162 measurement pole and is presented in Figure 2. The cloud optical and microphysical properties
163 were thus assessed by three independent instruments: a PMS Forward Scattering Spectrometer
164 Probe (FSSP-100), a Cloud Particle Imager (CPI) and a Polar Nephelometer (PN). They were
165 all connected to the same pump by plastic tubes, leading to the sampling volume indicated on
166 Figure 2. They were operated approximately 2 m above the platform level and mounted on a
167 tilting and rotating mast, allowing them to be moved manually in the prevailing wind direction.
168 The proper alignment of their inlet with the flow was based on the wind direction measurements
169 performed by a mechanical and ultrasonic anemometer.

170

171 The FSSP-100 measures the number and the size of particles going through the sampling
172 volume, from the forward scattering of a 632.8 nm wavelength laser beam (Knollenberg, 1981,
173 Dye and Baumgardner, 1984). Using the Mie theory, this instrument is dedicated to droplets.
174 The Particle Size Distribution (PSD) is thus computed in 15 adjustable size classes with
175 uncertainties on the effective diameter and *LWC* of respectively 2 μm and 30 % (Febvre et al.,
176 2012).

177 The CPI is an imager and takes pictures of the particles when going through the detection
178 volume with 256 grey levels, thanks to a CCD camera with a resolution of 1024×1024 pixels.
179 These images allow computing the particles size and so the PSD, but several morphological



180 parameters are also retrieved and are used to classify the sampled particle in 10 shape
181 categories: spheroid, needle, column, plate, bullet, stellar, graupel, rosette, sideplane and
182 irregular (Lefèvre, 2007). However, a manual classification has been done during the
183 CLIMSLIP campaign due to some malfunctions of the automatic classification. The
184 determination of the IWC is realized according to the Baker and Lawson (2006) and Lawson
185 and Baker (2006) method. The uncertainties on the concentration and the effective diameter are
186 assessed respectively as 50 % and 80 %.

187 The PN measures the scattering phase function of a set of cloud particles thanks to a 804 nm
188 wavelength laser beam and 56 photodiodes distributed over scattering angles between 3.5° and
189 172.5° (Gayet et al., 1997). From the scattering phase function can be computed two important
190 integrated optical parameters, the extinction coefficient and the asymmetry parameter with
191 accuracies estimated within 25% and 4%, respectively (Gayet et al., 2002).

192 The Nevzorov probe is a hot wire device at constant temperature with two captors and an
193 electrical resistor. The particles are vaporized, and an electrical power is provided to the
194 resistor. The resulting power is related to the *LWC* and *TWC*, depending on the captor
195 (Korolev et al., 1998). Due to high discrepancies, this instrument was used only for instrumental
196 comparison and data processing analysis and will not be discussed further.

197



198

199 Figure 2: Cloud instrumentation used during CLIMSLIP. Indicated are: particle size range,
200 main cloud properties measured and theoretical sampling speed.

201

202

203 2.2.2 Aerosol instrumentation

204

205 The particle inlet at the Mount Zeppelin station is a Whole Air Inlet, which possesses a heating
206 system that prevents the inlet to be filled by ice or frost and to evaporate the condensed water
207 or ice. Thus, all the aerosols (CCN, IN or interstitial) are sampled by the instruments described
208 hereafter. The aerosol sampling covers particles sizes between 3 and 809 nm (Tunved et al.,
209 2013).

210

211 The Mount Zeppelin aerosol instrumentation is composed of one Condensation Particle Counter
212 (CPC), one Differential Mobility Particle Sizer (DMPS), one aethalometer and one aerosol
213 nephelometer, which are running continuously throughout the year. The CPC, 3015A model, is
214 a particle counter for aerosol diameters larger than 3 nm. It measures aerosol concentration up
215 to 10⁵ particles/cm³ with an accuracy of 10 % (TSI, 2002). The DMPS is a CPC combined with
216 a Differential Mobility Analyzer (DMA), which allows selecting different size ranges. The
217 aerosol PSD is obtained with 22 diameter classes going from 25 to 809 nm. The aethalometer
218 assesses the Black Carbon (BC) concentration based to the optical extinction of the aerosols



219 collected on a filter (see Eleftheriadis et al., 2009, for details). The nephelometer measures the
220 aerosol scattering coefficient for three wavelength: 450, 550 and 700 nm (TSI, 2005). During
221 CLIMSLIP, this nephelometer was used with a time resolution of 5 minutes.

222

223 2.2.3 Data processing

224

225 The three cloud instruments operated at a one Hz resolution. The data processing has followed
226 the conclusions of the cloud instrumentation study presented in Guyot et al. (2015). This paper
227 highlights the biases that can exist between the instruments and the need of an Ensemble of
228 Particles Probe (EPP) to standardize the data. In the case of the CLIMSLIP campaign, such
229 correction was not possible for two reasons not developed further. (1) Strong discrepancies of
230 the EPP Nezhvorov probe, probably because of a too low sampling speed. (2) The
231 standardization according to the extinction coefficient of the PN is not consistent with the
232 aerosol data (there are more droplets than CCN). Thus, this study will not provide quantitative
233 results but qualitative ones based on case comparisons and variation studies.

234 According to Guyot et al. (2015), measurements with an angle between the instruments
235 orientation and the wind direction higher than 30° can modify the PSD due to changes in the
236 sampling conditions. Those measurements were therefore not taken into account for the study.
237 Moreover, the ground based low sampling speed induces low sampling rate, especially for the
238 CPI with values between 0.5 and 20 sampled particles per minute. This doesn't allow us to
239 work on low time resolution scale. To get sufficient particle statistics, the minimum average
240 time resolution will be 1 minute for the FSSP and one day for the CPI.

241

242 During the aircraft campaign, a cloud particle can break on impaction with the inlet due to the
243 high sampling speed corresponding to the plane speed. This results in more numerous and
244 smaller droplets or crystals and creates artifacts in the PSD (Rogers et al., 2006). Due to the
245 low sampling speed, ground based measurements has the advantage to avoid this effect, but at
246 the expense of the sampling rate.

247

248

249 **3 Identification and characterization of the study cases**

250

251 **3.1 Overview**

252

253

254 Several kinds of episodes were met during CLIMSLIP. Figure 3.a shows those episodes with
255 the time series of measured temperature and relative humidity. Thus, according to the
256 ceilometer measurements and observations, we enumerate:

- 257 - 4 episodes of sampling of the liquid and mixed phase layer (LMPL) of MPC, on March
258 11th and 29th and April 27th and 28th.
- 259 - 3 cases of sampling of the precipitation layer of MPC, on March 28th and April 14th and
260 20th.
- 261 - 2 occurrences of Blowing Snow (BS), on March 23th and 31th.

262

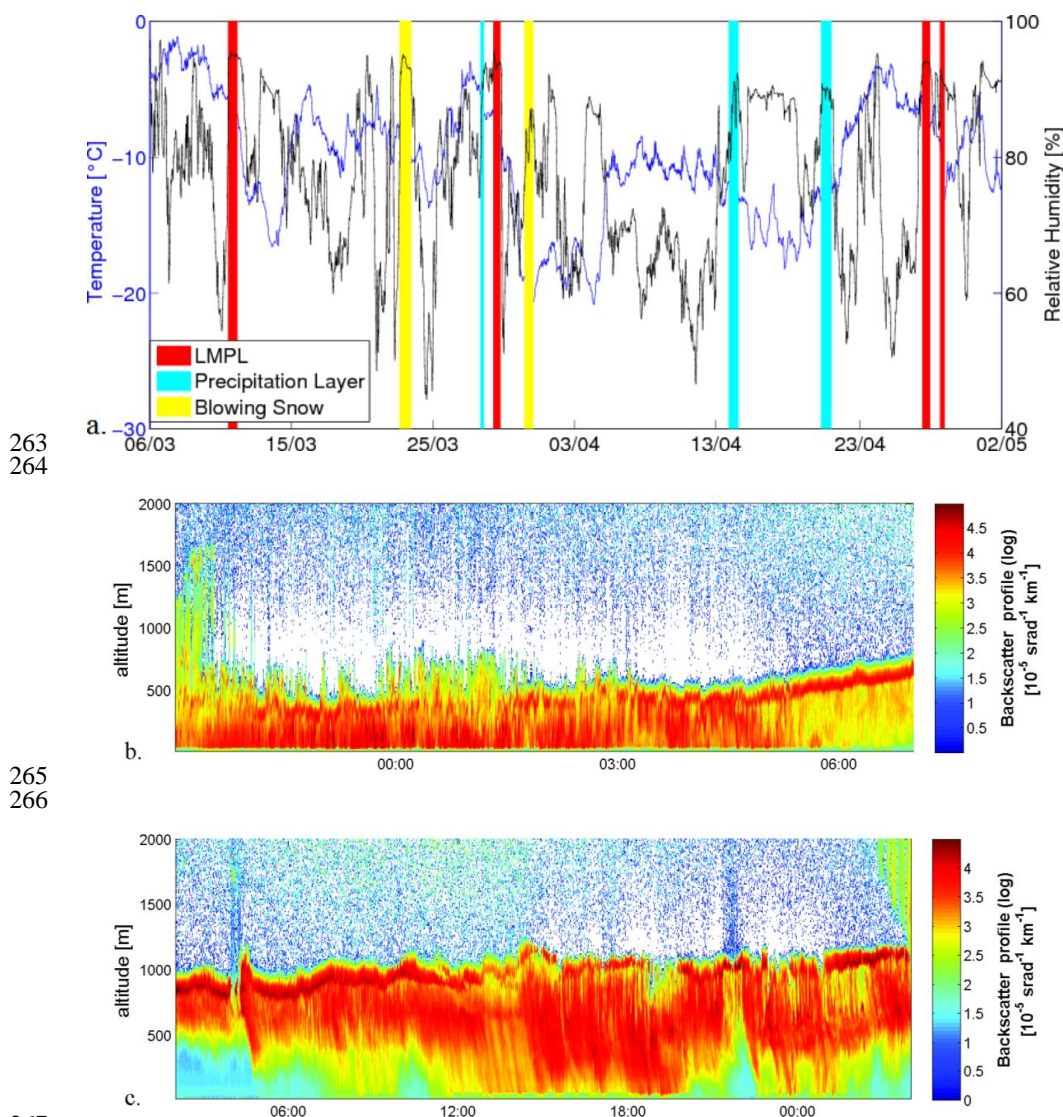


Figure 3: a) Time series of the temperature and relative humidity measured at the Zeppelin station. The different cases are plotted with colored columns. Examples of time series of the ceilometer attenuated backscattered coefficient profile in $\text{km}^{-1} \text{sr}^{-1}$ for b) MPC case (April 27th) and c) precipitation case (April 14th).

274 The cases called “LMPL” and “Precipitation layer” both reveal the presence of a MPC, i.e.
275 where cloud in situ instrumentation sampled ice and/or liquid particles. But, in the first case,
276 the ceilometer shows the liquid layer around 500 meters altitude (Figure 3.b). This liquid layer
277 having a very strong extinction coefficient, the ceilometer beam does not go through, what
278 happens above the liquid layer is therefore unknown. On the same time, droplets are sampled
279 by the FSSP. Following the altitude of the cloud, the station is in the liquid layer or the mixed
280 phase layer. These episodes are characterized by relative humidity maximum.



281 In the second case, the ceilometer locates the liquid layer around 1 km altitude or more (Figure
282 3.c). No droplets are sampled. The station is so below the mixed layer, within the ice
283 precipitation. This layer has a variable extinction coefficient depending on the crystal density
284 but the laser beam is not completely attenuated. The relative humidity shows high values around
285 90 % but remains lower than the MPC cases.

286 Moreover, the temperature varied between -20 to -1 °C, so it remains always below the
287 solidification point, liquid particles were always supercooled droplets. The Blowing Snow
288 episodes will be discussed in annex.

289

290 In the following, the LMPL and precipitation layer cases will be microphysically and optically
291 characterized. These characterizations will be useful to determine futures measurements that
292 are not completed with visual observations (e.g., remote sensing measurements). Moreover,
293 combined with other measurement campaign in the Arctic, we hope to increase knowledge
294 about growth processes in low level mixed phase arctic clouds.

295

296

297 **3.2 Characterization of the LMPL cases**

298

299 Arctic MPC can be characterized by a succession of layers with liquid or ice dominance. The
300 phase heterogeneity is both horizontal and vertical. Because of the fixed position of the
301 measuring station, we could not control the location of the measurements within the cloud
302 system. However, a characterization of the mean parameters is possible.

303

304 The determination of the thermodynamic phase of a cloud can be based on microphysical and
305 optical criteria. Figure 4 presents the occurrence number of the MPC liquid fraction F_{liq} and the
306 asymmetry parameter g . F_{liq} is computed as :

307

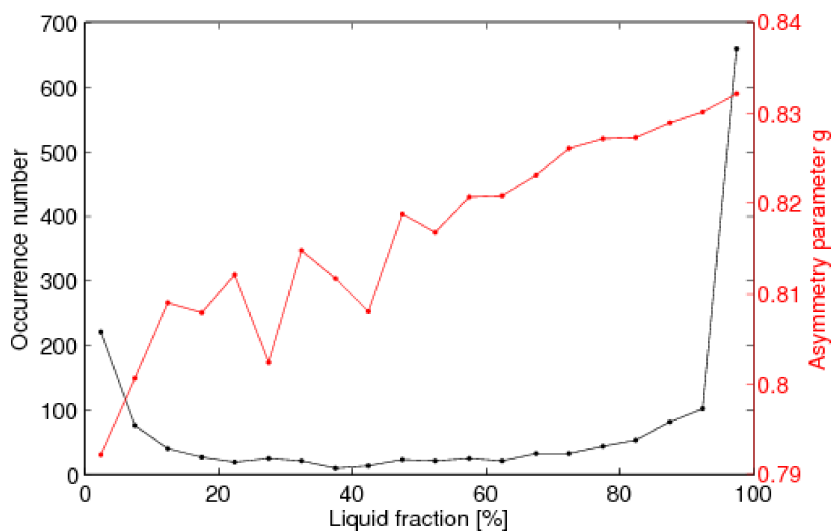
$$308 \quad F_{liq} = \frac{LWC_{FSSP}}{(LWC_{FSSP} + IWC_{CPI})} \quad (1)$$

309

310 The results show a higher observation frequency for extreme F_{liq} values (close to 0 or 100 %).
311 The minimum frequency is between 20 and 70 %. This means that the low level mixed phase
312 cloud layers are preferentially with liquid or ice dominance for the spatial resolution of our
313 measurements. This confirms the conclusions from the scientific literature (e.g., Gayet and al.,
314 2009; Korolev and Isaac, 2006).

315 Moreover, g shows a more or less linear relation with F_{liq} . This highlights the relation between
316 the optical properties and the microphysical properties. Therefore, the knowledge of the MPC
317 microphysical properties is a key parameter to reliably assess the radiative transfer in the Arctic.
318 The g variability is significantly larger for F_{liq} below 50 %. This tends to show a more complex
319 optical behavior for ice dominating layers.

320



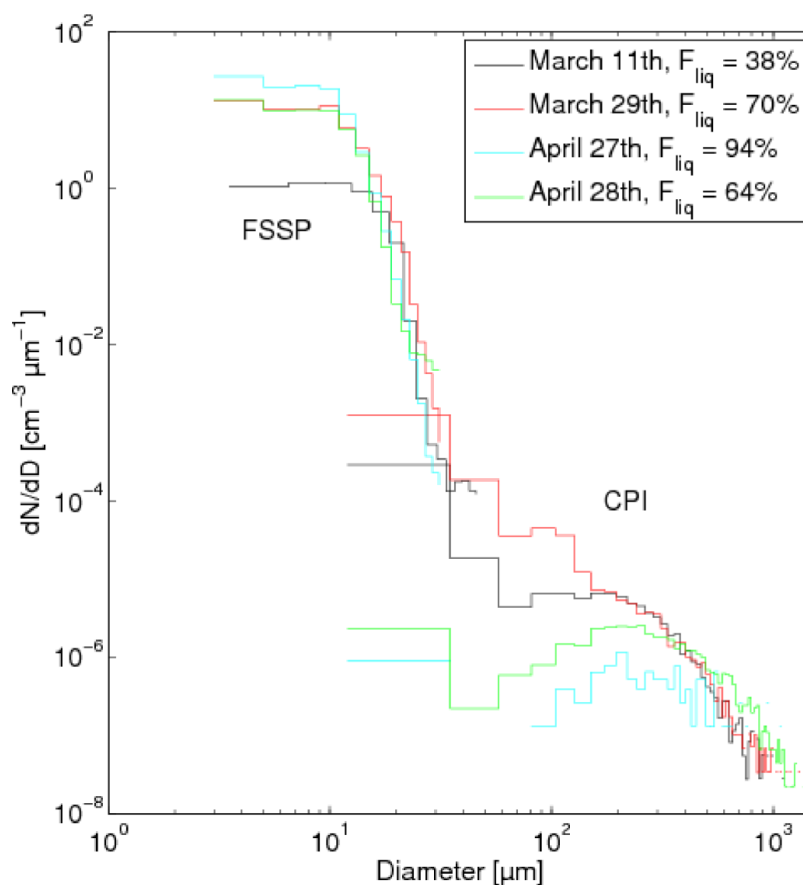
321
322 Figure 4: Occurrence number and mean values of g in relation to the liquid fraction F_{liq} for the
323 four LMPL cases. F_{liq} is derived from the CPI and FSSP measurements (see Eq. 1) with 1
324 minute resolution corresponding to a spatial resolution of 800 meters.

325
326
327 Figure 5 shows the average PSD, from 3 μm to 2.3 mm, obtained with the FSSP and the CPI
328 for the four LMPL cases. The mean F_{liq} is also indicated. The four PSD show similar trends,
329 i.e. two modes centered at 10 μm for droplets and around 250 μm for ice crystals.

330 According to Costa et al. (2014), these PSD correspond to the coexistence regime characterized
331 by RH_w (relative humidity according to liquid water) and RH_i (relative humidity according to
332 ice) $> 100\%$ and stable coexistence of crystals and supercooled liquid droplets with the droplet
333 PSD 10^6 higher than the crystal PSD. This is opposite to the Bergeron regime where $RH_w < 100\%$
334 and $RH_i > 100\%$, so the crystals grow in expense of the droplets (Costa et al., 2014). This
335 reveals that the Wegener-Bergeron-Findeisen process doesn't alone explain the formation and
336 growth of ice crystals.

337 However, the March 11th and 29th PSDs show differences with the other cases with a high
338 concentration for the smallest CPI classes. This is due to big droplets sampled by the CPI. The
339 FSSP doesn't show such consequent differences in droplet PSD or diameter. We also point out
340 that the absolute values should not be taken into account. Indeed, in addition to instrumental
341 issues (see Guyot et al., 2015), the results and the differences between the cases are largely
342 dependent on the station residence time within the liquid or mixed layer which cannot be
343 controlled. Similar PSDs were observed at the Mount-Zeppelin station by Uchiyama et al.
344 (2013) in 2011. This publication concludes that the liquid/ice distribution is a function of the
345 cloud evolution stage; we highlight here the importance of the station position inside the cloud
346 system for our data analysis.

347
348



349

350 Figure 5: Cloud PSD in $\text{cm}^{-3} \mu\text{m}^{-1}$ measured by the FSSP [3-50 μm] and the CPI [15-2300351 μm] and average for the four LMPL experiments. The mean values of F_{liq} are indicated in

352 legend.

353

354

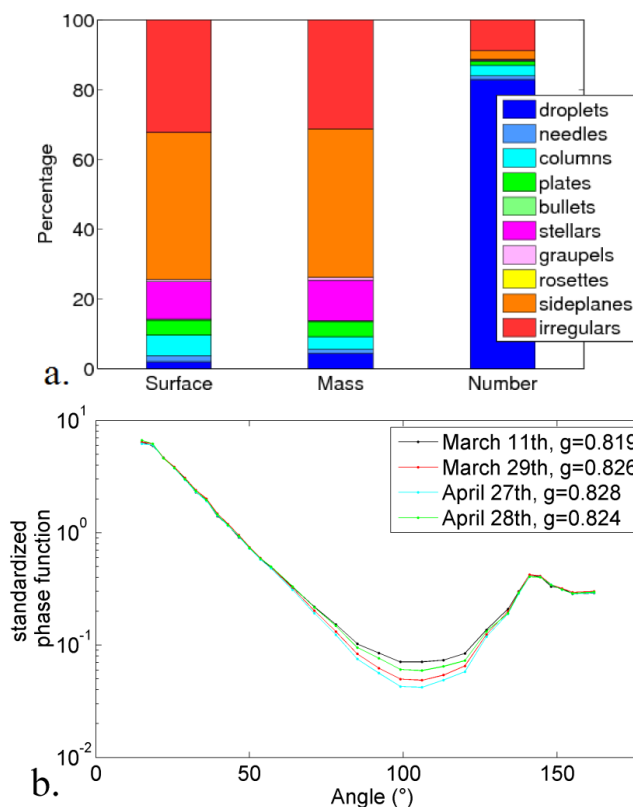
355 The shape classification performed by the CPI is presented Figure 6.a. The high droplet
356 concentrations of the smallest CPI classes observed on March 11th and 29th (see Figure 5) are
357 responsible of the strong number dominance of the droplets with a value of around 85 %.
358 However, liquid water represents a very small proportion in mass and surface fractions. For
359 these two quantities, side planes and irregular shapes dominate.

360 The assessment of the crystal growth mode is confronted to the fact that the measurement
361 station can change its position in the cloud. An evolution in the CPI PSD is so not necessary
362 due to particles growth. However, the crystal shape, accurately measured by the CPI, is a good
363 indicator for the growth mode and the high percentage of regular shape would indicate a growth
364 dominated by vapor deposition. Aircraft measurements performed in Svalbard in 2007 show
365 similar results, with in particular strong presence of irregulars and side planes for altitudes and
366 temperatures around 500 m and -12 °C, respectively (Gayet et al., 2009)

367



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Figure 6: a) CPI shape classification in surface, mass and number for the LMPL cases. The color represents the occurrence percentage of the shapes as indicated by caption, and b) average standardized phase functions measured by the PN for the four LMPL cases. Caption indicates the asymmetry parameter.

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Measurements of the cloud particle scattering properties performed by the PN allow to study the optical signature of the main microphysical properties observed on the MPC. Figure 6.b displays the average phase functions and asymmetry parameters (g) for the four LMPL cases. Differences between the experiments are negligible in forward and backward scattering but within the lateral scattering domain [60°; 130°]. The scattering increases when g decreases. The 1 minute average g values during the whole measurement campaign are included between 0.74 and 0.85, which is consistent with results obtained by Garrett et al. (2001).

Combined with Figure 5, these results show that g and the lateral scattering are related to microphysical properties. Indeed, lateral diffusion increases when F_{liq} decreases. Therefore, March 11th experiment presents the lower F_{liq} (38 %), the higher lateral scattering and the lower average value of g (0.819). The contrary is shown in the April 27th case ($F_{liq} = 94\%$, $g = 0.828$). This is consistent with previous studies (Gayet et al., 2009; Jourdan et al., 2010) and also proves the qualitative coherence between the FSSP and the PN.

The analysis of the optical-microphysics coupling is limited by the sampling speed and rate and the PN measurement accuracy. Indeed, a mean component analysis failed to establish a relationship between the phase function and the crystal morphology, as highlighted by Jourdan et al. (2010).



392 3.3 Characterization of the precipitation cases

393

394 Figure 7 displays the average CPI PSD for the three cases of precipitation. The FSSP is blind
395 for those particles sizes. The April 14th and 20th experiments show a PSD with very low
396 concentrations, close to the detection limit, centered around 350 μm and accompanied by
397 relatively low temperature $< -10^\circ\text{C}$. The March 28th case differs from the two other experiments
398 with higher concentrations and a PSD centered around 200 μm , similar to the LMPL cases.
399 Besides, the temperature is higher with an average value of -5°C . This could reveal an influence
400 of the mixed layer and/or temperature effect.

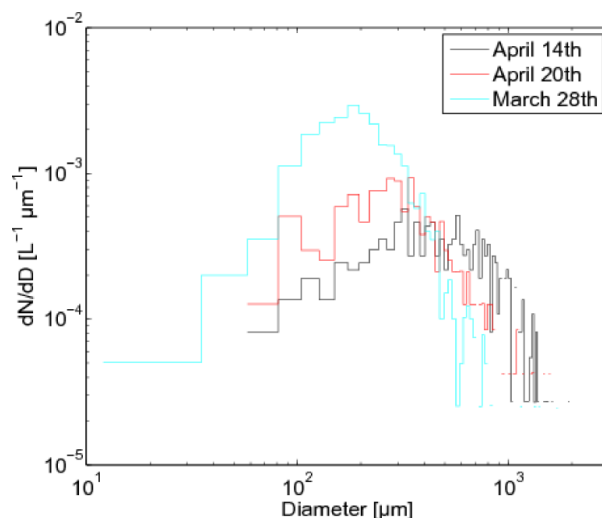
401 However, the ceilometer located the cloud base at an altitude of approximately 1000 m for the
402 three days, which would indicate that the station position doesn't explain the differences. The
403 temperature differences could lead to different growth processes and so different sizes.

404 This information can be provided by the CPI image classification presented Figure 8.a showing
405 a pronounced presence of stellars. Even if the stellar crystals aren't a majority in number, they
406 stand for more than half of total surface and mass. However, the number shape distribution was
407 not identical for the three days. Indeed, the 14th and 20th April experiments are dominated by
408 stellar whereas the March 28th case shows a much more important contribution of plaque,
409 irregular and needle. As the concentration is 5 times higher for the March 28th case, its
410 contribution in the total number distribution is more important.

411 Therefore, even if the temperature measured at the station is potentially different than the crystal
412 formation and growth temperatures and the oversaturation according to the ice was not
413 measured, we have seen that the CPI measurements show that temperatures below -10°C are
414 favorable to the formation of big size crystals such as stellar, whereas, for warmer temperatures,
415 plaque, irregular and needle crystals with smaller sizes dominate. This agrees with the crystal
416 classification studied by Bailey and Halley (2009) and explains the differences in the daily PSD.

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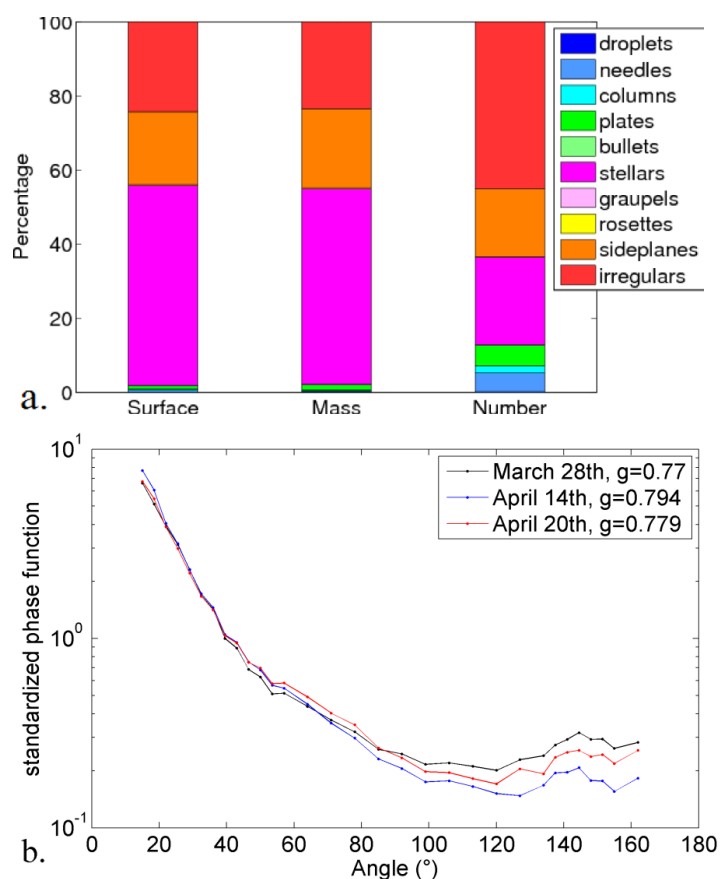


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Figure 7: Same as Figure 5 for the precipitation cases.



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427 Figure 8.b presents the average phase functions of the precipitation cases. The lateral scattering
 428 is more important than for the measurements of the mixed and liquid layers. The asymmetry
 429 parameter is lower around 0.79 which is typical for ice particles. Notable differences
 430 are observed for scattering angles as low as 40°. These differences are probably due to the crystal
 431 morphology variability. Unfortunately, such relationships were not observed with the
 432 CLIMSLIP data. Indeed, a principal component analysis didn't allow discriminating the phase
 433 function according to the crystal shape. This can be explained by the low sampling rate during
 434 the precipitation events involving a very low crystal statistics.

435

436

437 To conclude, the results were limited by the low particle sampling rate and the uncontrolled
 438 position of the station inside the cloud system. However, differences between LMPL and
 439 precipitation layer have been explicated and allow a quick recognition without visual
 440 observations in future studies. These results will be compared to other measurement campaign
 441 for a better understanding of the microphysical processes and feedbacks that take place in low
 442 level mixed phase arctic clouds.

443

444



445 **4 Aerosol-cloud interaction in the Arctic**

446

447 The objective of this part is to quantify the effects of the aerosol properties on the cloud
448 properties observed during the CLIMSLIP campaign. To do this, we will in a first step compare
449 the two experiments of March 11th and 29th that will be the “clean” and “polluted” cases,
450 respectively. In a second step, several aerosol cloud-interaction processes will be evaluated and
451 in situ measurements will be used to assess quantities that are required in parametrization of the
452 arctic aerosol-cloud interaction.

453

454 **4.1 Section on tools: the FLEXPART-WRF model and definitions**

455

456 This analysis will be supported by results from the lagrangian particle dispersion model
457 FLEXPART-WRF (version 3.1, Brioude et al., 2013) adapted from the FLEXPART model
458 (version 6.2, Stohl et al., 2005). FLEXPART-WRF simulates long distance transport and, in a
459 mesoscale, the moist and dry scavenging and the diffusion of atmospheric tracers and air masses
460 (see Stohl et al., 1998, Stohl and Thomson, 1999, or Stohl et al., 2005, for more details). The
461 FLEXPART-WRF model was driven by WRF (Skamarock et al., 2008) meteorological
462 forecasts to provide air masses back-trajectories and several tracers’ origins.

463

464 For each single run, 20000 pseudo-particles were released from a small volume surrounding
465 the analyzed position. Then, they were then tracked backward in time. The model output a
466 tridimensional distribution of the Potential Emission Sensitivity (PES) on a 1° longitude x 1°
467 latitude resolution grid. The PES is expressed in s/kg, which corresponds to the residence time
468 of air particles within a given cell. In order to investigate the potential sources of the pollution
469 transported to the Arctic and since the pollutants generally remain below the inversion layer,
470 the model output is integrated over the first kilometer atmospheric column and becomes a
471 Footprint PES (FPES). Combined with the ECLIPSE (Evaluating the Climate and Air Quality
472 Impacts of Short-Lived Pollutants, see Klimont et al., 2016) atmospheric pollutants emission
473 inventory, FLEXPART-WRF provides a valuable insight on the potential geographic
474 contribution of anthropogenic sources for pollution tracers such as CO, SO₂ and BC. The
475 combination between the FPES and the emissions is called the Potential Source Contribution
476 (PSC) expressed in kg of tracer per air kg. In this study, we will focus on the CO tracer which
477 gives an assessment on the origin of the anthropogenic pollution transported to Svalbard.

478

479 The aerosol cloud interaction study will also be supported by two other parameters: the
480 activation fraction F_a and the activation diameter D_a . F_a can be defined as the percentage of
481 aerosols becoming CCN (Abdul-Razzak et al., 1998) and is computed by the ratio of the FSSP
482 and CPC concentration. The CPC was chosen because it provides the largest aerosol size range:

483

$$484 \quad F_a = \frac{N_{drop}}{N_{aerosol}} = \frac{N_{FSSP}}{N_{CPC}} \quad (2)$$

485

486

487 We define D_a as the diameter beyond which all the aerosols are activated, assuming the aerosol
488 chemistry effect is negligible (Abdul-Razzak et al., 1998). During CLIMSLIP, D_a is calculated
489 as the DMPS diameter for which the DMPS total concentration is equal to the FSSP
490 concentration. Even if the aerosol size range is smaller for the DMPS than the CPC, the DMPS
491 was chosen because the aerosol PSD is necessary:

492



493

$$\sum_{D_{max}}^{D_a} n_{DMPS}(D) = N_{FSSP} \quad (3)$$

494

495 4.1 The “Clean” case of March 11th

496

497 The March 11th case, just like March 29th, presents a stable atmosphere with a low level mixed
 498 phase cloud. The liquid layer was sampled but, unfortunately, the ceilometer beam was almost
 499 entirely attenuated within the first 500 meters, avoiding to assess cloud top and base altitude.

500 The sounding balloon show the inversion layer around 925 mb (700 m) for both days.

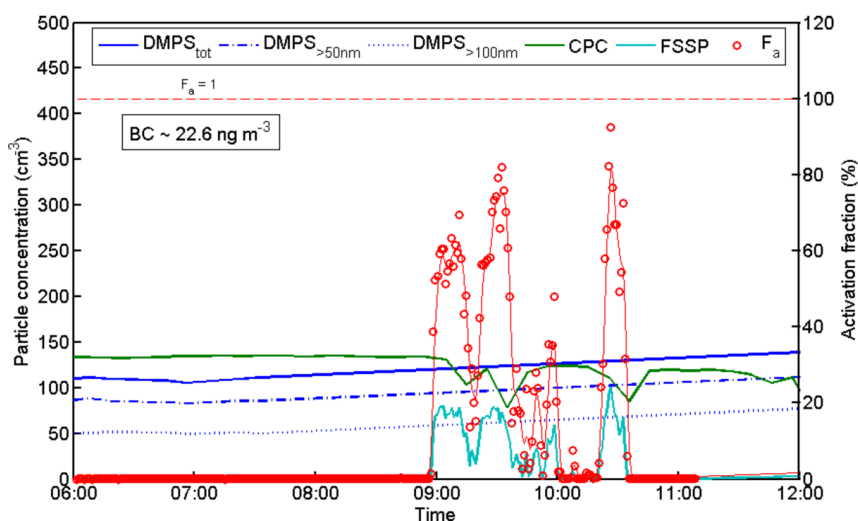
501 Figure 9 shows the time evolution of the DMPS, CPC and FSSP concentration, the activation
 502 fraction and the average BC concentration. The DMPS ceased to work from 7:30 until the
 503 following day, but, as the CPC and aethalometer parameters show almost constant values until

504 12:00, the DMPS concentration is assumed to do the same. The DMPS concentration is plotted
 505 for different particles sizes (total, > 50 nm and > 100 nm). The DMPS PSD shows a bimodal
 506 distribution with a pronounced Aitken mode which is as important as the accumulation mode
 507 (not shown). This is obvious in Figure 9 where the accumulation mode concentration, i.e.
 508 particles sizes larger than 100 nm, equals half the total concentration.

509 The CPC displays an aerosol concentration (> 3 nm) relatively stable and weak between 100
 510 and 130 cm⁻³. The average BC concentration reaches 22.6 ng m⁻³ during the liquid episode. The
 511 FSSP shows a droplet concentration up to 100 cm⁻³, which leads to F_a values between 60 and
 512 80 % for the sections clearly in the densest zone of the MPC liquid layer.

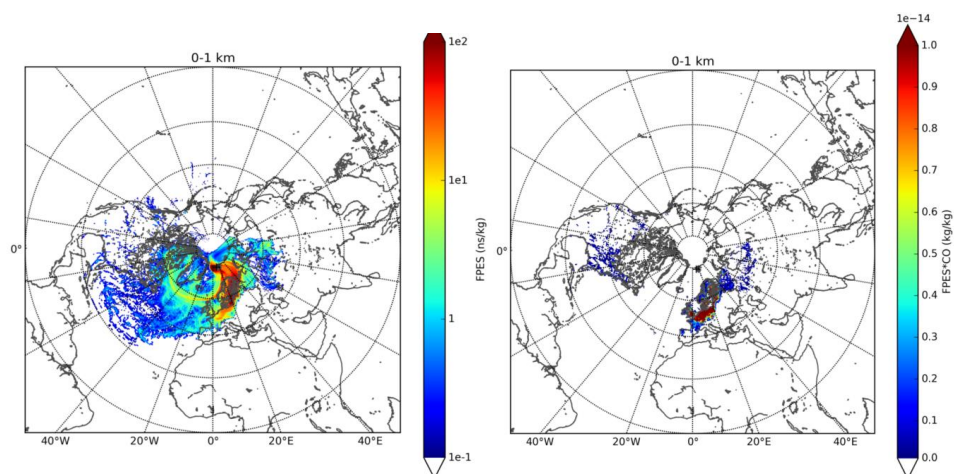
513 The FSSP droplet effective diameter is around 12 μm and the DMPS effective diameter around
 514 250 μm. D_a shows very high variations with a mean value around 150 nm (not shown).

515



516

517 Figure 9: Time series of aerosol concentrations measured by the DMPS and the CPC, droplet
 518 FSSP concentration and the activation fraction, for the March 11th. The DMPS concentration
 519 was divided into three groups: the total concentration [25 - 809 nm], particles larger than 50
 520 nm [50 - 809 nm] and larger than 100 nm [100 - 809 nm], the latter corresponding to the
 521 accumulation mode concentration. The activation fraction has been plotted with a sliding average
 522 of 5 minutes; the average aethalometre BC concentration is indicated.



523
524 Figure 10: a) FLEXPART-WRF 12 days FPES from the simulation initiated from Mount-
525 Zeppelin on March 11th between 9 AM and 12 AM UTC. b) PSC computed from the FPES of
526 Figure a) for the CO, expressed in kg CO per kg air.
527

528
529 Figures 10 a and b present respectively the FPES over 12 days and the PSC of CO for the air
530 mass arriving at the station during the liquid episode of the March 11th described in Figure 9.
531 The FPES shows that the aerosol sources are mainly located in the north of Scandinavia and so
532 that the long-range transport of anthropogenic aerosols is relatively limited. Indeed, over the
533 FLEXPART-WRF time computation of 12 days, the air masses come principally from Svalbard
534 and Scandinavia surrounding, showing very slow move. The CO PSC map presents an
535 anthropogenic origin dominated by North Europa: Scandinavia, north of Germany, Netherland,
536 Belgium and north of France.

537 The closer air masses origin makes this case the “clean” case. The important contribution of
538 local aerosol sources, mainly composed of gaseous precursors for the arctic region during this
539 period of the year (Quinn et al., 2007), explains the relative small aerosol mean diameter and
540 the high Aitken mode concentration observed by the DMPS (see Figure 9).
541

542

543 4.2 The “Polluted” case of March 29th

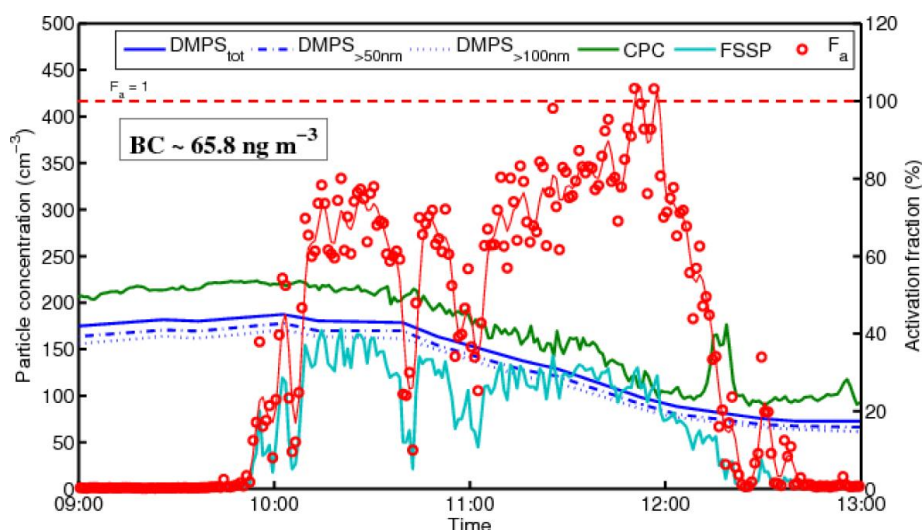
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545 Figure 11 displays the same time series as Figure 9 for the liquid episode of March 29th. The
546 CPC and DMPS total concentration are decreasing going respectively from 220 cm⁻³ to 120 cm⁻³
547 and from 175 cm⁻³ to 80 cm⁻³, due to the scavenging by ice precipitation. The FSSP droplet
548 concentrating reaches 150 cm⁻³ and the average BC concentration 65.8 ng m⁻³. Comparing to
549 the March 11th case, these four concentration are all higher during the March 29th. The activation
550 fraction is also higher on March 29th with values between 80 and 100 % in the liquid layer and
551 F_a increases as the aerosol concentration decreases.

552 Just like March 11th, D_a shows very variable values around 150 nm (not shown). Its high
553 variability makes this parameter unsuitable when comparing the two cases. However, D_a
554 decreases from around 150 nm to 50 nm and the FSSP effective diameter increases from 8 to
555 10 μm when F_a increases, proving that smaller aerosol particles are activated and droplets grow
556 when the aerosol number decreases.



557 Moreover, the DMPS PSD shows that 90 % of the aerosol concentration is included in the
558 accumulation mode, with an effective diameter almost constant at 300 nm. Therefore, the
559 aerosol diameter is larger and the droplet diameter is smaller for the March 29th case compared
560 to the March 11th case.
561



562
563 Figure 11: Same as Figure 9 for the March 29th case
564

565 The differences observed between the two days can be explained by the air masses origin.
566 Figure 12 shows the same FLEXPART-WRF FPES and CO PSC for the air mass arriving at the
567 station during the liquid episode of the March 29th. Backtrajectories distinguish clearly two
568 origin regions. The first one is Western Europa. The second air mass shows higher values of
569 time residence and comes from northeast Asia: northeast China and extreme east Russia. The
570 particularity of March 29th consists thus in this air mass coming from Asia which is the region
571 generally accepted to emit the highest aerosol concentration compared to the others regions of
572 the world (Boucher et al., 2013).

573 Therefore, compared to the “clean” case of March 11th, March 29th shows long range transport
574 of anthropogenically influenced air masses, leading to higher aerosol concentration in the Arctic
575 with especially a BC mass concentration 3 times higher. Thus, March 29th constitute the
576 “polluted” case. According to Quennehen et al. (2012), during the route, the Aitken mode
577 concentration quickly decreases by coagulation, for the benefit of the accumulation mode,
578 increasing the average effective diameter. This explains the accumulation mode dominance
579 observed in Figure 11 and the increase of the average DMPS effective diameter, and confirms
580 the strong influence of the lower latitudes emissions during the “polluted” case. On the contrary,
581 the “clean” case shows local sources composed of fresh particles, for at least half the
582 concentration.

583 This long range anthropogenic pollution has also strong influence on cloud properties. Indeed,
584 CCN abilities being mainly due to the aerosol size in the Arctic (Mc Farquhar et al., 2011),
585 accumulation mode dominance leads to higher aerosol effective diameter and higher F_a values.
586 Combined with higher aerosol concentration, the droplet concentration increases whereas the
587 droplet size decreases meaning, theoretically, that the cloud optical thickness increases.
588

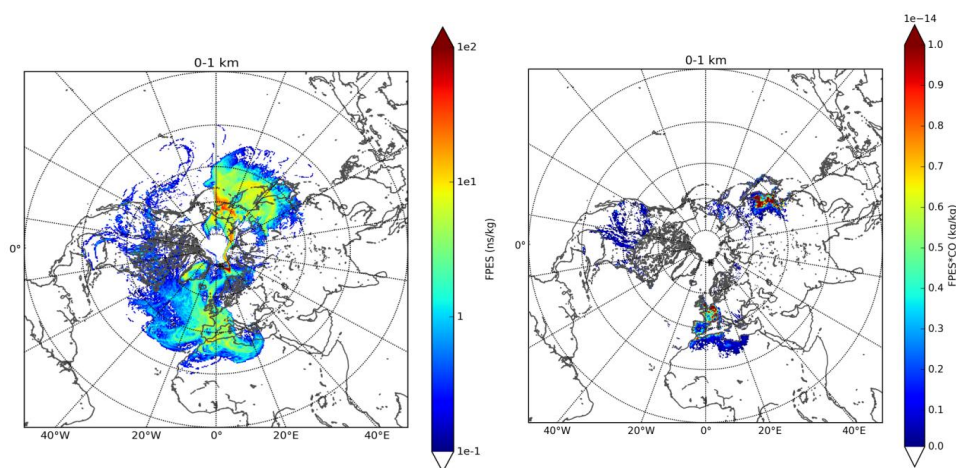


Figure 12: a) and b) Same as Figure 10 for the air masses arriving in the Mount-Zeppelin station on March 29th between 10 AM and 1 PM UTC.

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This qualitative study has to be completed with quantitative parameters that can be found in the scientific literature. Therefore, the next section will focus on the quantitative variations of droplet concentration and size according to aerosol properties. Moreover, glaciation and riming indirect effect will be assessed.

598 4.3 Quantification of the impacts of the aerosol properties on the cloud 599 microphysical properties

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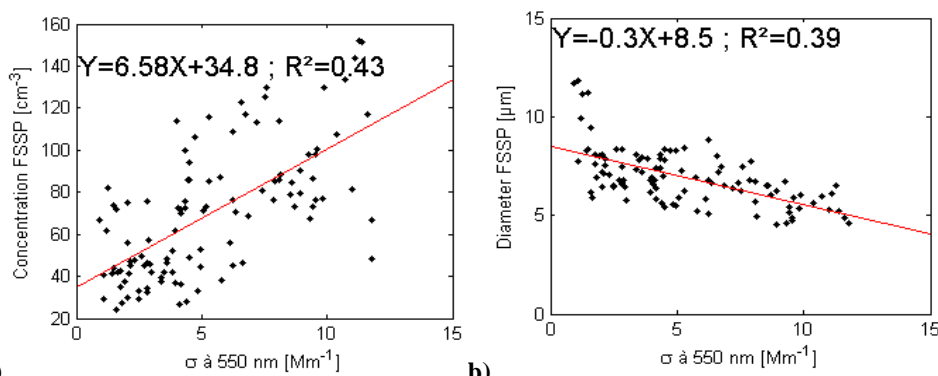
The sensitivity of cloud diameter and concentration according to aerosol haze will be assessed from two parameters, called the Indirect Effect parameter (*IE*) and the Nucleation Efficiency (*NE*) and defined as follows (Feingold et al., 2001, 2003, Garrett et al., 2004):

$$607 \quad IE = - \frac{\partial \ln(r_e)}{\partial \ln(\sigma)} \quad (4)$$

$$608 \quad 609 \quad NE = \frac{\partial \ln(N)}{\partial \ln(\sigma)} \quad (5)$$

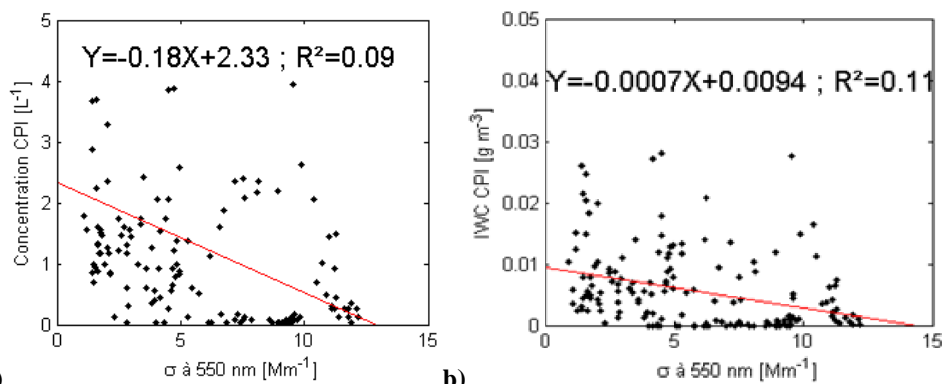
610 where r_e is the droplet effective radius, N the droplet concentration and σ the aerosol scattering
 611 coefficient.

612 We made two assumptions to use these parameters. First, *IE* and *NE* are assumed to evaluate
 613 the variations of the droplet concentration and size according to the CCN concentration. To
 614 measure this one, we use the scattering coefficient which is assumed to be proportional to the
 615 CCN concentration. The accumulation mode particles are the most inclined to serve as CCN
 616 because of their size and possess the highest scattering cross section compared to the other
 617 modes (Garrett et al., 2004). Second, r_e and N are also dependent on the *LWP*, so *IE* and *NE*
 618 have to be computed for similar *LWP* clouds (Feingold et al., 2001). During CLIMSLIP, the
 619 *LWP* was not measured and we assumed that the *LWP* is effectively constant. This is reasonable
 620 since the sampled clouds were all low level mixed phase arctic clouds and from the same season.
 621



622 **a)**
 623 **b)**
 624 Figure 13: Comparison of the aerosol scattering coefficient σ at $\lambda = 550$ nm measured by the
 625 nephelometer, with a) the droplet concentration, and b) the effective diameter measured by
 626 the FSSP, for the four LMPL cases. Values of LWC below $5 \cdot 10^{-3} \text{ g m}^{-3}$ were not taken into
 627 account. This comparison has been performed with a nephelometer time resolution of 5
 628 minutes.

629 Figure 13 presents the comparisons between the droplet concentration and diameter with the
 630 aerosol scattering coefficient. The results are consistent with the Twomey effect (Twomey,
 631 1974, 1977) and the Albrecht effect (Albrecht, 1989). The correlation coefficients R^2 are equal
 632 to 0.43 and 0.39 for concentration and size, respectively. This high dispersion can be explained
 633 by the fact that the droplet concentration and size depend also of LWP , temperature and the
 634 position of the measurement volume within the cloud system.
 635 At $\lambda = 550$ nm, $IE = 0.2$ and $NE = 0.43$ were obtained. This is to compare with the study of
 636 Garrett et al. (2004) performed at Barrow, Alaska, where IE and NE were found to be between
 637 0.13 and 0.19 and between 0.3 and 0.5 respectively, at $\lambda = 550$ nm. Very similar values are so
 638 found in two different regions of the Arctic, which confirms these parameterizations of the first
 639 and second aerosol indirect effect for the arctic region.
 640



641 **a)**
 642 **b)**
 643 Figure 14: 5 minutes comparison of the aerosol scattering coefficient σ at $\lambda = 550$ nm with a)
 644 the concentration and b) the IWC of crystals sampled by the CPI, for the four LMPL cases. A
 645 threshold of $50 \mu\text{m}$ was applied to the particles diameter to discard the droplets sampled by
 646 the CPI.
 647



648 The glaciation (Lohmann, 2002a, 2002b) and the riming indirect effect (Borys et al., 2003) were
649 evaluated during the CLIMSLIP campaign thanks to the CPI and the nephelometer
650 measurements. The comparison between the crystal concentration and *IWC* with σ (or nuclei
651 concentration) is displayed in Figure 14. The results show very weak correlation, for the
652 concentration and the *IWC*. This means that neither the glaciation nor the riming indirect effect
653 were revealed during CLIMSLIP.

654 This absence can be due to the high uncertainty in the CPI measurements and/or to low sampling
655 rate that leads to a very low statistical representation. To compare, the study of Jackson et al.
656 (2012), during the ISDAC and MPACE campaign, found a correlation corresponding to the
657 glaciation effect above the cloud liquid phase but no evidence of the riming effect.

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696 **5 Summary and conclusion**

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698 Within the framework of the arctic amplification, the complex interactions between the cloud
699 and aerosol properties remain a challenge to enhance the arctic cloud modeling and to get a
700 better quantification of the consequences of the anthropogenic pollution on the arctic climate.
701 The ANR project CLIMSLIP (CLimate IMPacts of Short-Lived Pollutants in the polar region)
702 provides new data from a ground based aerosol and cloud instrumentation located at the Mount
703 Zeppelin station, Ny-Alesund, Svalbard, during spring 2012. This instrumentation contains a
704 FSSP, a CPI and a Polar Nephelometer to sample clouds and a CPC, a DMPS and a
705 nephelometer for aerosols.

706 During the campaign, four cases of LMPL (Liquid and Mixed Phase Layer), three cases of snow
707 precipitation layer and two cases of BS (Blowing Snow) were sampled. The precipitation layer
708 cases correspond to the lower layer of a MPC. The precipitation events are composed of large
709 crystals (Mean Diameter $D_m \sim 350 \mu\text{m}$) with an important presence of stellar. The LMPL events
710 are characterized by a bimodal PSD with a large number of droplets. The liquid mode was
711 located around $10 \mu\text{m}$ and the crystal mode around $250 \mu\text{m}$. The phase function measurements
712 showed an increase of the lateral scattering as F_{liq} decreases.

713 According to Guyot et al. (2015), only isoaxial measurements with a wind speed higher than 5
714 m/s are selected. This deleted a non-negligible amount of data and so limited the analysis,
715 especially for the precipitation cases where the particle statistics were the weaker. Moreover,
716 the position of the station within the cloud system was approximate despite the ceilometer
717 measurements.

718 A study by comparison of the effects of the anthropological aerosols transported to the Arctic
719 was performed. According to the FLEXPART/WRF simulations, the “polluted” case of March
720 29th showed air masses from Europe and East Asia whereas the aerosol sources during the
721 “clean” case of March 11th were closer (mainly from Scandinavia) and the anthropogenic
722 contribution doesn't exceed northern Europe.

723 Thus, the polluted case presents higher Black Carbon, aerosol and droplets concentrations, a
724 more important accumulation mode, smaller droplet sizes and higher activation fraction F_a . The
725 March 29th activation diameter D_a decreased when the droplet diameter increased and F_a
726 increased, proving that smaller aerosol particles are activated and droplets grow up when the
727 aerosol number decreases. These results confirm the first and second aerosol indirect effects
728 with the coefficients IE and NE respectively around 0.2 and 0.43. These values are very close
729 to those found by Garrett et al. (2004), which performed measurement at Barrow in Alaska, and
730 are so good candidates to be used to parameterize arctic aerosol-cloud interaction in climate
731 models. Furthermore, the crystal concentration and IWC do not show any correlation with the
732 aerosol properties, which indicates that the glaciation and riming indirect effects are not
733 highlighted during the CLIMSLIP-NyA campaign.

734

735

736 *Acknowledgements.* This work was supported by the ANR project CLIMSLIP and the conseil
737 general de l'Allier. We also thank the AVI for providing the ceilometer data and the ITM and
738 NILU for monitoring the Mount Zeppelin station. We are grateful to scientists, engineers and
739 technicians that make this study possible. Boris Quennehen acknowledges the IPSL
740 CICLAD/CLIMSERV mesocenter for providing computing resources.

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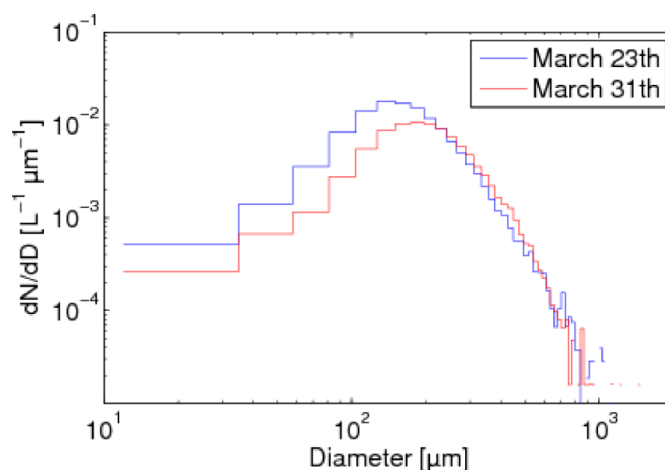
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746 **Annex: Characterization of the Blowing Snow (BS) cases**

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748 During ground based measurements, some snow was collected that was suspended in the
749 atmosphere due to wind. This is the so-called Blowing Snow (BS). This annex aims at the
750 microphysics characterization in order to recognize this kind of episode and the optical
751 properties measurements, especially the phase function, that can be used as a reference to
752 develop new parameterizations of the snow simple scattering properties (Räsänen et al., 2015).
753



754 Figure 15: Same as Figure 5 for the BS cases.

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When the BS occurs, the sky is clear as observed by the ceilometer. However, crystal particles are sampled. They are snowflakes initially resting on the ground but getting suspended in the air by the wind.

Figure 15 shows the average PSDs measured by the CPI for the two BS cases. The shape and the amplitude are similar for the two PSDs, with a mean diameter between 150 and 200 μm . for a maximum class concentration around $10^{-2} \text{ L}^{-1} \mu\text{m}^{-1}$. The CPI shape classification, plotted in Figure 16.a, shows a large prevalence of irregular crystals, as well in number, surface or mass (i.e. volume), with a percentage around 90% of the crystals. These two characteristics constitute the microphysics signature of the BS. The difference between the BS and MPC (see Figure 5 and 6) signature makes it possible to identify BS events even if the station is located inside a cloud.



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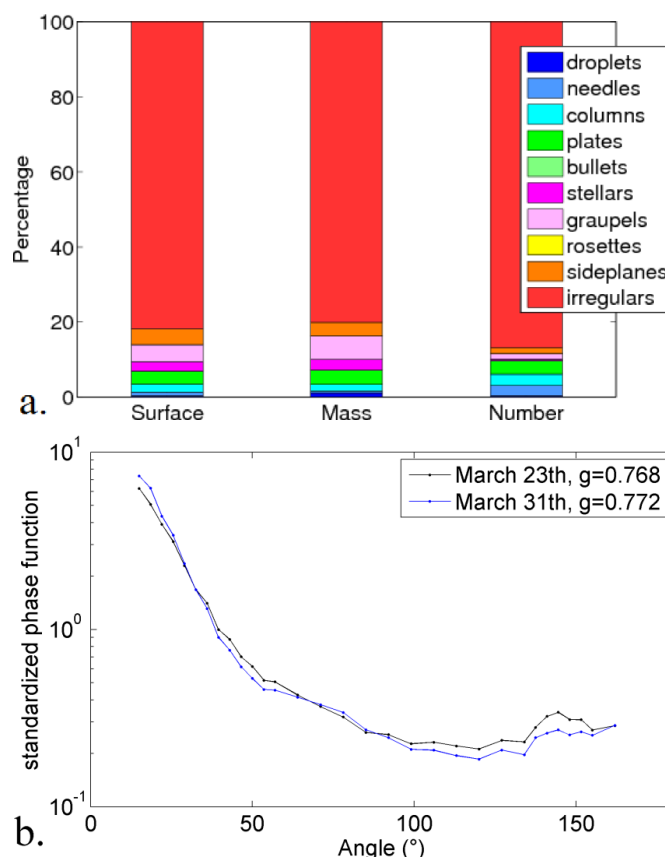


Figure 16: Same as Figure 6 (a and b) for the BS cases.

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Even if the resuspension of crystals in the air can modify the shape by impacts, we consider that the sampled crystals are similar to the deposited precipitations and aged for several days. Thus, the BS events during CLIMSLIP were excellent occasions to measure the arctic snow properties.

Figure 16.b displays the average phase functions of the BS cases. The shape of the curves are very similar to the precipitation cases, typical of ice particles, but with lower g values. These measurements constitute a unique database to develop parameterizations of the arctic snow optical properties. Indeed, in most of the climate and weather forecast models, the computation of the snow albedo uses the approximation of spherical snow grain (Wang and Zeng, 2010). Thus, the study of Räisänen et al. (2015) proposes new modeling parameterizations of the snow single scattering properties (SSP) based on the CLIMSLIP-NyA in situ measurement of the phase function. The obtained snow SSP takes into account the complex BS particles shape (Räisänen et al., 2015).

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