



1	Supercooled liquid water clouds observed over Dome C,
2	Antarctica: temperature sensitivity and surface radiation impact
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# 23 Abstract

24	Clouds affect the Earth climate with an impact that depends on the cloud nature (solid/
25	liquid water). Although the Antarctic climate is changing rapidly, cloud observations are sparse
26	over Antarctica due to few ground stations and satellite observations. The Concordia station is
27	located on the East Antarctic Plateau (75°S, 123°E, 3233 m above mean sea level), one of the
28	driest and coldest places on Earth. We used observations of clouds, temperature, liquid water
29	and surface radiation performed at Concordia during 4 austral summers (December 2018-2021)
30	to analyse the link between liquid water and temperature and its impact on surface radiation in
31	the presence of supercooled liquid water (liquid water for temperature less than 0°C) clouds
32	(SLWCs). Our analysis shows that, within SLWCs, temperature logarithmically increases from
33	-36.0°C to -16.0°C when liquid water path increases from 1.0 to 14.0 g m <sup>-2</sup> , and SLWCs
34	positively impact the net surface radiation, which logarithmically increases by $0.0$ to $50.0$ W
35	$\rm m^{-2}$ when liquid water path increases from 1.7 to 3.0 g m^{-2}. We finally estimate that SLWCs
36	have a great potential radiative impact over Antarctica whatever the season considered, up to
37	5.0 W m <sup>-2</sup> over the Eastern Antarctic Plateau and up to 30 W m <sup>-2</sup> over the Antarctic Peninsula
38	in summer.





#### 40 **1. Introduction**

41 Antarctic clouds play an important role in the climate system by influencing the Earth's radiation balance, both directly at high southern latitudes and, indirectly, at the global level 42 43 through complex teleconnections (Lubin et al., 1998). However, in Antarctica, ground stations 44 are mainly located on the coast and yearlong observations of clouds and associated 45 meteorological parameters are scarce. Meteorological analyses and satellite observations of 46 clouds can nevertheless give some information on cloud properties suggesting that clouds vary 47 geographically, with a fractional cloud cover ranging from about 50 to 60% around the South 48 Pole to 80-90% near the coast (Bromwich et al., 2012; Listowski et al., 2019). In situ aircraft 49 measurements performed mainly over the Western Antarctic Peninsula (Grosvenor et al., 2012; 50 Lachlan-Cope et al., 2016) and nearby coastal areas (O'Shea et al., 2017) provided new insights 51 to polar cloud modelling and highlighted sea-ice production of Cloud-Condensation Nuclei and 52 Ice Nucleating Particles (see e.g. Legrand et al., 2016). Mixed-phase clouds (made of solid and 53 liquid water) are preferably observed near the coast (Listowski et al., 2019) with larger ice crystals and water droplets (Lachlan-Cope, 2010; Lachlan-Cope et al., 2016; Grosvenor et al., 54 55 2012; O'Shea et al., 2017; Grazioli et al., 2017). Based on the raDAR/liDAR-MASK 56 (DARDAR) spaceborne products (Listowski et al., 2019), it has been found that clouds are 57 mainly constituted of ice above the continent whereas the abundance of Supercooled Liquid 58 Water (SLW, the water staying in liquid phase below 0°C) clouds depends on temperature and 59 liquid/ice fraction, decreases sharply poleward, and is two to three times lower over the Eastern 60 Antarctic Plateau than over the Western Antarctic. An important point remains the inability of 61 both research and operational weather prediction models to accurately represent the clouds (especially SLW clouds, SLWCs) in Antarctica causing biases of several tens W m<sup>-2</sup> on net 62 63 surface radiation (Listowski and Lachlan-Cope, 2017; King et al., 2006, 2015; Bromwich et al., 64 2013) over and beyond the Antarctic (Lawson and Gettelman, 2014; Young et al. 2019). From





65 year-long LIDAR observations of mixed-phase clouds at South Pole (Lawson and Gettelman,

66 2014), SLWCs were shown to occur more frequently than in earlier aircraft observations or

67 weather model simulations, leading to biases in the surface radiation budget estimates.

68 Liquid water in clouds may occur in supercooled form through heterogeneous nucleation due to a relative lack of ice nuclei for temperature greater than -39°C and less than 0°C. Very 69 70 little SLW is then expected because the ice crystals that form in this temperature range will 71 grow at the expense of liquid droplets (called the "Wegener-Bergeron-Findeisen" process; 72 Storelymo and Tan, 2015). Nevertheless, SLW is often observed at negative temperatures 73 higher than -20°C at all latitudes being a danger to aircraft since icing on the wings and airframe 74 can occur, reducing lift, and increasing drag and weight. As temperature decreases to -36°C, 75 SLW dramatically lessens, so it is highly difficult 1) to observe SLWCs and 2) to quantify the 76 amount of liquid water present in SLWCs. But during the Year Of Polar Prediction (YOPP) 77 international campaign, recent observations performed at the Dome C station in Antarctica of 78 2 case studies in December 2018 have revealed SLWCs with temperature between -20°C and -79 30°C and Liquid Water Path (LWP, the liquid water concentration integrated along the vertical) 80 between 2 to 20 g m<sup>-2</sup>, as well as a considerable impact on the net Surface Radiation (SR) that 81 exceeded the simulated values by 20-50 W m<sup>-2</sup> (Ricaud et al., 2020).

82 The Dome C (Concordia) station, jointly operated by French and Italian institutions in the 83 Eastern Antarctic Plateau (75°06'S, 123°21'E, 3233 m above mean sea level, amsl), is one of 84 the driest and coldest places on Earth with surface temperatures ranging from about -20°C in summer to -70°C in winter. There are three main instruments relevant to this study that have 85 86 been routinely running for about 10 years: 1) The H<sub>2</sub>O Antarctica Microwave Stratospheric and 87 Tropospheric Radiometer (HAMSTRAD, Ricaud et al., 2010a) to obtain vertical profiles of temperature and water vapour, as well as the LWP. 2) The tropospheric depolarization LIDAR 88 89 (Tomasi et al., 2015) to obtain vertical profiles of backscatter and depolarization to be used for





(1)

- 90 the detection of SLWCs. 3) The Baseline Surface Radiation Network (BSRN) station to
  91 measure surface longwave (4–50 μm) and shortwave (0.3–3 μm), downward and upward
  92 radiation from which the Net Surface Radiation (Net SR), calculated as the difference between

the downward and upward SRs, can be computed (Driemel et al., 2018) as:

- 94 Net SR = LWD SR LWU SR + SWD SR SWU SR
- 95 where LWD SR, LWU SR, SWD SR and SWU SR correspond to Longwave Downward,
- 96 Longwave Upward, Shortwave Downward and Shortwave Upward SRs, respectively.
- 97 The article is structured as follows. Section 2 presents the instruments during the period of 98 study. In section 3, we detail the methodology employed to detect the SLWCs and calculate 99 their impact on SR, and we present the statistical method to emphasize the relationship between 100 temperature and LWP on one hand, and SR and LWP on the other hand. The results are 101 highlighted in section 4 and discussed in section 5, before concluding in section 6.

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#### 103 2. Instruments

We have used the observations from 3 instruments held at the Dome C station, namely the LIDAR instrument to classify the cloud as SLWC, the HAMSTRAD microwave radiometer to obtain vertical profiles of temperature and Liquid Water Path (LWP) and the BSRN network to measure the Surface Radiation (SR) components: LWD, LWU, SWD and SWU to finally obtain the Net SR.

109 2.1. LIDAR

The tropospheric depolarization LIDAR (532 nm) has been operating at Dome C since 2008 (see http://lidarmax.altervista.org/englidar/\_Antarctic%20LIDAR.php). The LIDAR provides 5-min tropospheric profiles of aerosols and clouds continuously, from 20 to 7000 m above ground level (agl), with a resolution of 7.5 m. LIDAR depolarization (Mishchenko et al., 2000) is a robust indicator of non-spherical shape for randomly oriented cloud particles. A





- 115 depolarization ratio below 10% is characteristic of SLWC, while higher values are produced by
- 116 ice particles. The possible ambiguity between SLWC and oriented ice plates is avoided at Dome
- 117 C by operating the LIDAR 4° off-zenith (Hogan and Illingworth, 2003).
- 118 2.2. HAMSTRAD

119 HAMSTRAD is a microwave radiometer that profiles water vapour, liquid water and 120 tropospheric temperature above Dome C. Measuring at both 60 GHz (oxygen molecule line 121 (O<sub>2</sub>) to deduce the temperature) and 183 GHz (H<sub>2</sub>O line), this unique, state-of-the-art 122 radiometer was installed on site for the first time in January 2009 (Ricaud et al., 2010a and b). 123 The measurements of the HAMSTRAD radiometer allow the retrieval of the vertical profiles 124 of water vapour and temperature from the ground to 10-km altitude with vertical resolutions of 125 30 to 50 m in the Planetary Boundary Layer (PBL), 100 m in the lower free troposphere and 126 500 m in the upper troposphere-lower stratosphere. The time resolution is adjustable and fixed 127 at 60 seconds since 2018. Note that an automated internal calibration is performed every 12 128 atmospheric observations and lasts about 4 minutes. Consequently, the atmospheric time 129 sampling is 60 seconds for a sequence of 12 profiles and a new sequence starts 4 minutes after 130 the end of the previous one. The temporal resolution on the instrument allows for detection and 131 analysis of atmospheric processes such as the diurnal evolution of the PBL (Ricaud et al., 2012) 132 and the presence of clouds and diamond dust (Ricaud et al., 2017) together with SLWC (Ricaud 133 et al., 2020). In addition, the LWP (g m<sup>-2</sup>) that gives the amount of liquid water integrated along 134 the vertical can also be estimated. Observations of LWP have been performed when the 135 instrument was installed at the Pic du Midi station (2877 amsl, France) during the 136 calibration/validation period in 2008 prior to its set up in Antarctica in 2009 (Ricaud et al., 2010a) and during the Year Of Polar Prediction (YOPP) campaign in summer 2018-2019 137 138 (Ricaud et al., 2020). At the present time, it has not yet been possible to compare HAMSTRAD 139 LWP retrievals with observations from other instruments, neither at the Pic du Midi nor at





Dome C stations. To better evaluate its performance, the 2021-2022 and the future 2022-2023 summer campaigns are dedicated to in-situ observations of SLWCs. Comparisons with numerical weather prediction models were showing consistent amounts of LWP at Dome C when the partition function between ice and liquid water was favouring SLW for temperatures less than 0°C (Ricaud et al., 2020).

145 2.3. BSRN

The BSRN sensors at Dome C are mounted at the Astroconcordia/Albedo-Rack sites, with upward and downward looking, heated and ventilated Kipp&Zonen CM22 pyranometers and CG4 pyrgeometers providing measurements of hemispheric downward and upward broadband shortwave (SW,  $0.3-3 \mu m$ ) and longwave (LW,  $4-50 \mu m$ ) fluxes at the surface, respectively. These data are used to retrieve values of net surface radiation. All these measurements follow the rules of acquisition, quality check and quality control of the BSRN (Driemel et al., 2018).

152 2.4. Period of study

153 From the climatological study presented in Ricaud et al. (2020), the SLWCs are mainly 154 observed above Dome C in summer, with a higher occurrence in December than in January: 155 26% in December against 19% in January representing the percentage of days per month that 156 SLW clouds were detected during the YOPP campaign (summer 2018-2019) within the LIDAR 157 data for more than 12 hours per day. We have thus concentrated our analysis on December and 158 the 4 years: 2018-2021. Since we have to use the three data sets (LIDAR, HAMSTRAD and 159 BSRN) in time coincidence, the actual number of days per year selected in our analysis is 160 presented in Table 1.

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# 165 3. Methodology

# 166 3.1. SLWC detection and Surface Radiation Impact

Consistent with Ricaud et al. (2020), we use LIDAR observations to discriminate between 167 SLW and ice in a cloud. High values of LIDAR backscatter ( $\beta > 100 \beta_{mol}$ , with  $\beta_{mol}$  the 168 169 molecular backscatter) associated with very low depolarization ratio (< 5%) signifies the 170 presence of a SLWC whilst high depolarization ratio (>20%) indicates the presence of an ice 171 cloud or precipitation. Once the SLWC is detected both in time and altitude, the temperature  $(\theta)$  profile within the cloud and the LWP measured by the HAMSTRAD radiometer in time 172 coincidence are selected together with the SR observed by the BSRN instruments: Net, LWD, 173 LWU, SWD and SWU SR. Figure 1 shows, as a typical example, the time evolution of the 174 175 LIDAR backscatter and depolarization ratio, the HAMSTRAD LWP and temperature vertical 176 profile for the 27 December 2021. Associated with these SLWCs, the LWP increases with time from 1.0 to 3.0 g m<sup>-2</sup>. The SLWCs are present over a temperature range varying from about -177 28.0 °C to -33.0 °C. Note the cloud present at 04:00-05:00 UTC that is not labelled as a SLWC 178 179 but rather as an ice cloud (high backscatter and high depolarization signals) with no associated 180 increase of LWP and temperature above -28.0 °C.

Figure 2 highlights the time evolution of the SLWC obtained on 27 December 2021 together with some snapshots from the HALO-CAM video camera taken with or without SLWC on: 01:00 (no SLWC), 07:19 (SLWC), 09:00 (no SLWC), 10:14 (SLWC), 13:00 (no SLWC), 16:03 (SLWC), 18:01 (no SLWC) and 20:53 UTC (SLWC). SLWCs (high backscatter and low depolarization signals) are clearly detected at 07:00-08:00, 10:00-11:00, 16:00-17:00, 21:00-22:00 and 23:00-24:00 UTC over an altitude range 500-1000 m above ground level (agl). In general, SLWCs observed over the station did not correspond to overcast conditions.

188 Over the 4 summers (December 2018-2021), we have selected 3 datasets in time 189 coincidence with SLWC: LWP,  $\theta$  and SR. In order to estimate the impact of the SLWC onto





190 the SR, we calculated the anomaly of the daily SR with respect to the clear-sky SR associated 191 to the same day. Since it is impossible to measure for the same day the SR with and without cloud, we have considered, over the 4-summer period, clear-sky 24-hour periods. Only 5 clear-192 193 sky days were selected on: 2 and 19 December 2018, and 3, 17 and 26 December 2021. We 194 have considered these 5 days as the reference SRs  $(SR_{Ref})$  presented Figure 3. We have also 195 calculated (Figure 4) the time evolution of the clear-sky surface radiation variability ( $\delta SR_{Ref}$ ), 196 namely the difference in SR observed by the BSRN instruments between each of 5 clear-sky 197 day and the corresponding values averaged over the 5 days, for Net, LWD, LWU, SWD and 198 SWU SR. The  $SR_{Ref}$  for the 5 days shown Figure 3 are all consistent to each other with an 199 obvious diurnal cycle in Net, LWD, LWU, SWD and SWU SR (we recall that in December 200 there is a 24-h solar illumination at Dome C). The variability within the 5 days ( $\delta SR_{Ref}$ ) shown 201 Figure 4 is within ±20 W m<sup>-2</sup> for the Net SR with a greater Net SR in 2018 than in 2021, within ±35 W m<sup>-2</sup> for LWD and LWU SR (maxima on 17 December 2021 and minima on 2 December 202 2018), and within ±25 W m<sup>-2</sup> for SWD and SWU SR (maxima on 26 December 2021 and 203 204 minima on 2 December 2018).

Based on these 5  $SR_{Ref}$ , we performed a systematic study over the 4-summer period by calculating the surface radiation anomaly  $\Delta SR$  defined as:

 $\Delta SR = SR - SR_{Ref} \tag{2}$ 

for Net, LWD, LWU, SWD and SWU. As an example, we show in Figure 5 the time evolution for 27 December 2021 of the presence of the SLWC together with  $\Delta SR$  calculated with respect to  $SR_{Ref}$  set to 26 December 2021. Associated with the SLWC, LWD and LWU  $\Delta SR$  increase by 30-50 and 10-30 W m<sup>-2</sup>, respectively, whilst SWD and SWU  $\Delta SR$  decrease by 50-150 W m<sup>-2</sup> , respectively. The effect on the Net  $\Delta SR$  is positive (10-100 W m<sup>-2</sup>) at 07:00-08:00, 10:00-11:00, 16:00-17:00 UTC and negative (from -30 to -100 W m<sup>-2</sup>) at 21:00-22:00 and 23:00-24:00 UTC. We thus want to statistically analyse all the  $\Delta SR$  calculated in 2018-2021 with the 5  $SR_{Ref}$ 





- 215 in order to check whether the net effect of the SLWC on the SR is positive or negative and to
- 216 evaluate its sensitivity to liquid water amounts.
- 217 3.2. Statistical Method

218 The datasets are binned into 1°C-wide bins for  $\theta$ , 0.2 g m<sup>-2</sup>-wide bins for LWP, and 5 W 219 m<sup>-2</sup>-wide bins for  $\Delta SR$ . The number of points per bin is calculated for all the paired datasets, 220 namely  $\theta$ -LWP, and  $\Delta SR$ -LWP (Net  $\Delta SR$ -LWP, LWD  $\Delta SR$ -LWP, LWU  $\Delta SR$ -LWP, SWD 221  $\Delta SR$ -LWP and SWU  $\Delta SR$ -LWP). The 2D distribution of the Probability Density Function (PDF) is calculated for the paired datasets and defined as  $PDF_{ij} = 100 \frac{N_{ij}}{N_t}$ , where  $N_{ij}$  and  $N_t$ 222 are the count number in the bin *ij* and the total count number  $(N_t = \sum_{j=1}^N \sum_{i=1}^M N_{ij})$ , 223 224 respectively, with M and N being the total number of bins in LWP on one side, and in 225 temperature or  $\Delta SR$  on the other side, respectively. For each value of  $\theta_i$  within a 1°C-wide bin *j* or  $\Delta SR_i$  within a 5 W m<sup>-2</sup>-wide bin *j*, a weighted average of LWP ( $\overline{LWP_1}$ ) is calculated together 226 227 with its associated weighted standard deviation ( $\sigma_{LWP_i}$ ) considering all the LWP<sub>ij</sub> values within 228 0.2 g m<sup>-2</sup>-wide bins from i=1 to M, with M the total number of LWP bins and  $w_{ij}$  the weight, 229 namely the number of points  $(w_{ij} = N_{ij})$ , associated to the bin *ij*:

$$\overline{LWP_j} = \frac{\sum_{i=1}^{M} w_{ij} \, \frac{LWP_{ij}}{\sum_{i=1}^{M} w_{ij}} \tag{3}$$

231 and

232 
$$\sigma_{LWP_j} = \sqrt{\frac{\sum_{i=1}^{M} w_{ij} \left(LWP_{ij} - \overline{LWP_j}\right)^2}{\sum_{i=1}^{M} w_{ij}}}$$
(4)

For each  $\theta$  and  $\Delta SR$  dataset, the distribution of the total count numbers  $N_{tj}$  per 1°C or 5 W m<sup>-</sup> <sup>2</sup>-wide bin ( $N_{tj} = \sum_{i=1}^{M} N_{ij}$  with j = 1, ..., N) can be fitted by a function N(x), with  $x = \theta$  or  $\Delta SR$ , based on 2 to 3 Gaussian distributions as:

236 
$$N(x) = \sum_{k=1}^{2 \text{ or } 3} a_k \exp\left(-\frac{1}{2} \left(\frac{x-\mu_k}{\sigma_k}\right)^2\right) + c$$
(5)





237	with $a_k$ , $\mu_k$ and $\sigma_k$ being the amplitude, the mean and the standard deviation of the $k^{\text{th}}$ Gaussian
238	function ( $k = 1, 2 \text{ or } 3$ ) and $c$ is a constant. We have used $k = 2$ for $\Delta SR$ and $k = 3$ for $\theta$ . Table
239	2 lists all the fitted parameters ( $a_k$ , $\mu_k$ , $\sigma_k$ and $c$ with $k = 1$ to 2 or 3).
240	In the relationship between $x (\theta \text{ or } \Delta SR)$ and LWP, we have considered $x_j (\theta_j \text{ or } \Delta SR_j)$ to
241	be significant when:
242	$x_j \le \mu_k \pm \sigma_k \text{ for } k = 1 \text{ or } 2 \text{ (for } \Delta SR) \text{ or } 3 \text{ (for } \theta)$ (6)
243	and highlighted this significant point by showing the associated $\overline{LWP_j}$ and $\sigma_{LWP_j}$ with $j = 1,,$
244	<i>N.</i>
245	Finally, a logarithmic function of the form
246	$x = a + b \ln(\overline{LWP}) \tag{7}$
247	has been fitted onto these significant points $(\overline{LWP_j} \pm \sigma_{LWP_j}, x_j)$ where the retrieved constants
248	<i>a</i> and <i>b</i> are shown in Table 3 for <i>x</i> being $\theta$ , Net $\Delta SR$ , LWD $\Delta SR$ , LWU $\Delta SR$ , SWD $\Delta SR$ and
249	SWU $\Delta SR$ .
250	
251	4. Results
252	4.1. Temperature-Liquid Water Relationship in Supercooled Liquid Water Clouds
253	The relationship between temperature and LWP within SLWCs over the 4-summer period
254	at Dome C is presented Figure 6 left in the form of a Probability Density Function (PDF) that
255	is the fraction of points within each bin of 0.2 g $m^{\text{-}2}$ width in LWP and 1.0°C width in
256	temperature. It clearly shows a net tendency for liquid water to increase with temperature, up
257	to ~14 g m <sup>-2</sup> in LWP and -18°C in temperature, with two zones having a density as high as
258	~2%, at [0.5 g m <sup>-2</sup> , -33°C] and [1.5 g m <sup>-2</sup> , -32°C]. Performing a weighted average of the LWPs
259	within each temperature bin (Figure 6 centre), fitting a Gaussian distribution of the count
260	numbers as a function of temperature (Figure 6 right) and considering only temperature bins
261	within one-sigma of the centre of the Gaussian distributions, we can fit the following





262 logarithmic relation of the temperature  $\theta$  as a function of LWP within the SLWC (Figure 6 263 centre):

264	$\theta(LWP) = -33.8 (\pm 1.5) + 6.5 \ln(LWP) \tag{8}$
265	for $\theta \in [-36; -16]$ °C and <i>LWP</i> $\in [1.0; 14.0]$ g m <sup>-2</sup> , where (±1.5) corresponds to the range
266	where the relationship is valid within the 2 blue dashed lines in Figure 6 centre. In other words,
267	based on our study, we have a clear evidence that supercooled liquid water exponentially
268	increases with temperature. Although the amount of LWP is very low ( $\ll 20 \text{ g m}^{-2}$ ) at Dome C
269	compared to what can be measured and modelled (Lemus et al., 1997) in the Arctic (50-75 g
270	m <sup>-2</sup> ) and at middle/tropical latitudes (100-150 g m <sup>-2</sup> ), we intended to estimate its impact on the
271	SR at Dome C.
272	4.2. Impacts of Supercooled Liquid Water Clouds on Surface Radiation

273 In Figures 7 to 9, the left panel presents the PDF (for bins of 0.2 g m<sup>-2</sup> width in LWP and 274 5 W m<sup>-2</sup> width in  $\Delta SR$ ) of the surface radiation anomaly  $\Delta SR$  as a function of the LWP, for Net, 275 LWD, LWU, SWD and SWU, respectively. The central panel shows, for the same parameters, 276 the corresponding weighted average LWP within 5 W m<sup>-2</sup>-wide bins of radiation anomaly 277 whereas the right panel shows the corresponding count number within 5 W m<sup>-2</sup>-wide bins.

278 Based on our analysis, the relationship between Net  $\Delta SR$  (in W m<sup>-2</sup>) and the LWP (in g m<sup>-2</sup>)

<sup>2</sup>) has been estimated from the HAMSTRAD and BSRN data as:

280  $Net \Delta SR(LWP) = -50.0 (\pm 10.0) + 90.0 \ln(LWP)$  (9)

for  $Net \Delta SR \in [-15; 50]$  W m<sup>-2</sup> and  $LWP \in [1.5; 3.0]$  g m<sup>-2</sup>, where (±10.0) corresponds to the range where the relationship is valid within the 2 blue dashed lines in Figure 7 centre. Thus, for LWP greater than 1.7 g m<sup>-2</sup>, our study clearly shows that there is a positive impact of SLWC on the Net  $\Delta SR$  that can reach 50 W m<sup>-2</sup> for an LWP of 3.0 g m<sup>-2</sup>.

The splitting of the net radiation anomaly between each of its four components can be evaluated from their individual relationships with the LWP. These relations are gathered in





287	Table 3, established from the plots presented in Figures 7 to 9. They are of the same form as
288	for net surface radiation anomaly, i.e. a logarithmic dependence on LWP. Table 3 presents the
289	coefficients <i>a</i> and <i>b</i> of the logarithmic function $f(LWP) = a + b \ln(LWP)$ for the temperature
290	$\theta$ or the radiation components $\Delta SR$ , together with the valid range of these relations for $\theta$ , $\Delta SR$
291	and LWP. For the values presented in Table 3, our study clearly shows that SLWCs have a
292	positive impact on LWD and LWU, with $\Delta SR$ increasing from 0 to 100 W m <sup>-2</sup> and from 0 to
293	40 W m <sup>-2</sup> for LWP ranging from 1.0 to 4.0 and from 1.6 to 2.5 g m <sup>-2</sup> , respectively.
294	Furthermore, our study also shows that SLWCs have a clear negative impact on SWD and
295	SWU, with $\Delta SR$ decreasing from 0 to -140 W m <sup>-2</sup> and from 0 to -75 W m <sup>-2</sup> with LWP ranging
296	from 1.2 to 3.8 and from 1.2 to 3.2 g m <sup>-2</sup> , respectively.

297

#### 298 5. Discussion

### 299 5.1. Logarithmic Dependency

300 It is interesting to note that the relationships we have obtained in our study showing an 301 exponential dependence of LWP on both temperature and SR anomaly is very similar to the 302 dependence of the molar volume and density of water on critical temperature. As a matter of 303 fact, the density  $\rho$  (g cm<sup>-3</sup>) and molar volume  $\nu$  (cm<sup>3</sup> mol<sup>-1</sup>) of liquid water are exponentially 304 varying with temperature (Sippola and Taskinen, 2018):

305 
$$\rho = \rho_0 \exp\{-T_c(A + B\varepsilon + 2C\varepsilon^{1/2})\}$$
(10)

306 
$$\nu = \frac{M_{H_2O}}{\rho} = \frac{M_{H_2O}}{\rho_0} \exp\{T_c(A + B\varepsilon + 2C\varepsilon^{1/2})\}$$
(11)

307 where  $\rho_0$  (g cm<sup>-3</sup>), A (K<sup>-1</sup>), B (K<sup>-1</sup>), and C (K<sup>-1</sup>) are parameters;  $T_c$  is the critical temperature 308 whose value varies from 227 to 228 K, and  $M_{H_20}$  (g mol<sup>-1</sup>) is the molecular weight of water. 309  $\varepsilon$  (unitless) is defined as:

$$\varepsilon = \frac{T}{T_c} - 1 \tag{12}$$





- 311 where *T* is temperature in K.
- 312 5.2. Modelling SLWC

313 Previous studies have already underlined the difficulty to model the SLWC together with 314 its impact on surface radiations. Modelling SLWCs over Antarctica is challenging because 1) 315 operational observations from meteorological radiosondes are scarce since the majority of 316 ground stations are located at the coast and very few of them are maintained all year long, and 317 satellite observations are limited to 60°S in geostationary orbit whilst, in polar orbit, the number 318 of available orbits does not exceed 15 per day, and 2) the model should provide a partition 319 function favouring liquid water at the expense of ice for temperatures between -36°C and 0°C in order to calculate realistic SLW concentrations. Differences of 20 to 50 W m<sup>-2</sup> in the Net SR 320 321 were found in the Arpege model (Pailleux et al., 2015) between clouds made of ice or liquid 322 water during the summer 2018-2019 (Ricaud et al., 2020), differences that are very consistent 323 with the results obtained in the present study. Although SLWCs are less present over the 324 Antarctic Plateau than over the coastal region, their radiative impact is not negligible and should 325 be taken into account with great care in order to estimate the radiative budget of the Antarctic 326 continent in one hand, and, on the other hand, over the entire Earth.

327 5.3. Errors

328 Measurements of temperature, LWP, depolarization signal and SR are altered by random 329 and systematic errors that may affect the relationships we have obtained between LWP and 330 either temperature or SR anomalies. The temperature measured by HAMSTRAD below 1 km 331 has been evaluated against radiosonde coincident observations from 2009 to 2014 (Ricaud et 332 al., 2015) and the resulting bias is 0-2°C below 100 m and between -2 and 0°C between 100 333 and 1000 m. SLWCs are usually located around 400-600 m above the ground where the cold 334 bias can be estimated to be about -1.0°C. The one-sigma (1- $\sigma$ ) root mean square (RMS) temperature error over a 7-min integration time is 0.25°C in the PBL and 0.5°C in the free 335





336 troposphere (Ricaud et al., 2015). As a consequence, given the number of points used in the 337 statistical analysis (>1000), the random error on the weighted-average temperature is negligible  $(<0.02^{\circ}C)$ . The LWP random and systematic errors are difficult to evaluate since there is no 338 339 coincident external data to compare with. Nevertheless, the 1- $\sigma$  RMS error over a 7-min integration time can be estimated to be 0.25 g m<sup>-2</sup> giving a random error on the weighted average 340 341 LWP less than 0.08 g m<sup>-2</sup>. Based on clear-sky observations, the positive bias can be estimated 342 to be less than 0.4 g m<sup>-2</sup>. Theoretically, SLW should not exist at temperatures less than -39°C 343 although it has been observed in recent laboratory measurements down to -42.55°C (Goy et al., 2018). Using equation (8) with an LWP bias of 0.4 g m<sup>-2</sup> gives a temperature of -39.8°C (~0.8°C 344 345 lower than the theoretical limit of -39°C), so the biases estimated for temperature and LWP are 346 very consistent with theory.

347 The estimation of systematic and random errors on LIDAR backscattering and 348 depolarization signals and their impact on the attribution/selection of SLWC is not trivial. But 349 the most important point is to evaluate whether the observed cloud is constituted of purely liquid 350 or mixed-phase water. Even considering the backscatter intensity only, we could not exclude 351 that ice particles could have been present in the SLWC events investigated in 2018 (Ricaud et 352 al., 2020). Therefore, in the present analysis, although we made a great attention to diagnose 353 ice in the LIDAR cloud observations, we cannot totally exclude ice particles thus mixed-phase 354 parcels were actually present when we labelled the observed cloud as SLWCs.

The 4 instruments providing LWD, LWU, SWD and SWU SR follow the rules of acquisition, quality check and quality control of the BSRN (Driemel et al., 2018). In polar regions (Lanconelli et al., 2011), the global SWU and SWD SRs are expected to be affected by random errors up to  $\pm 20$  W m<sup>-2</sup> while LWD SRs are expected to be affected by random errors not greater than  $\pm 10$  W m<sup>-2</sup> (Ohmura et al., 1998). As a consequence, given the large number





of observations used per 5 W m<sup>-2</sup>-wide bins (1000-3000), the random error on the weighted-360 average SRs is negligible (0.3-0.7 W m<sup>-2</sup>) whatever the radiations considered, LW and SW. 361 362 Finally, apart from the instrument-related SR<sub>Ref</sub> error, another source of error comes from 363 1) the geometry of observation and 2) the discontinuous SLWC layer. Firstly, LIDAR is almost zenith pointing, HAMSTRAD makes a scan in the East direction (from 10° elevation to zenith), 364 365 whilst the BSRN radiometers detect the radiation in a  $2\pi$ -steradians field of view (3D 366 configuration). That is to say, in our analysis, the whole sky contributes to the radiation whilst 367 only the cloud at zenith (1D configuration) and on the East direction (2D configuration) is observed by the LIDAR and HAMSTRAD, respectively. Secondly, SLWCs cannot be 368 369 considered as uniform in the whole (see e.g. broken cloud fields in Figure 2).

#### 370 5.4. Other clouds

371 Although the method we have developed to select the SLWCs has been validated using the 372 amount of LWP and, in another study, using space-borne observations (Ricaud et al., 2020), we 373 cannot rule out that, associated with the SLW droplets, are also ice particles, that is clouds are 374 constituted of a mixture of liquid and solid water. Generally, such clouds are a superposition of 375 a lower layer being made of liquid water and an upper layer being made of solid water (see Fig. 376 12.3 from Lamb and Verlinde, 2011). These mixed-layer clouds do not significantly modify the 377 relationship between temperature and LWP because 1) SLW observations from HAMSTRAD 378 are only sensitive to water in liquid phase and 2) temperature from HAMSTRAD is selected at 379 times and vertical heights where the LIDAR depolarization signal is very low (<5%). Although 380 we have verified that pure ice clouds were not selected by our method, we cannot differentiate 381 mixed-phase clouds from purely SLWCs. As a consequence, the relationship between  $\Delta SR$  and 382 LWP might be affected by the presence of mixed-phased clouds in addition to SLWCs. This 383 may explain the negative part of the Net, LWD and LWU  $\Delta SR$  ([-20, 0] W m<sup>-2</sup>) and the





384 positive part of the SWD and SWU  $\Delta SR$  ([0; 10] W m<sup>-2</sup>) for low values of LWP ([0.8; 1.6] g

385 m<sup>-2</sup>).

386 Furthermore, we already have noticed that SLWCs developed at the top of the PBL (Ricaud 387 et al., 2020) in the "entrainment zone" and maintained in the "capping inversion zone", 388 following the terminology of Stull (2012), at a height ranging from 100 to 1000 m above ground 389 level. Nevertheless, during the local "night" at 00:00-06:00 LT, the PBL may collapse down to 390 a very low height ranging 20-50 m. In this configuration, it is hard to differentiate from LIDAR 391 observations between a SLWC and a fog episode, although the LIDAR can measure 392 depolarization (but not backscatter) down to approximately 10-30 m above the ground (Figure 393 S3 in Chen et al., 2017), so that we can distinguish liquid/frozen clouds very close to the ground. 394 Finally, we cannot rule out that, above the SLWCs that are actually observed by both 395 LIDAR and HAMSTRAD, other clouds might be present, as e.g. cirrus clouds constituted of 396 ice crystals. These mid-to-upper tropospheric clouds cannot be detected by HAMSTRAD (no 397 sensitivity to ice crystals). In the presence of SLWCs either low in altitude or optically thick, 398 the LIDAR backscatter signal is decreased in order to avoid saturation and the signal from upper 399 layers is thus almost cancelled. These mid-to-high-altitude clouds are observed by the BSRN 400 instruments and SR can be affected in this configuration. Based on the presence of cirrus clouds 401 before or after the SLWCs (and sometimes during the SLWCs if optically thin), we can estimate 402 that the number of days when SLWCs and cirrus clouds are simultaneously present to cover 403 less than 10% of our period of interest.

404 5.5. Potential radiative impact of SLWCs over Antarctica

Based on our results and on the observed cloud fraction ( $\eta_{CF}$ ) of SLWCs over Antarctica for different seasons (Listowski et al., 2019), we have estimated the potential radiative impact of SLWCs at the scale of the Antarctic continent (*Net*  $\Delta SR_{global}^{max}$ ) from the maximum of *Net*  $\Delta SR$ (*Net*  $\Delta SR^{max} = 50$  W m<sup>-2</sup>) computed in our study:





$SR^{max}$ (13)	409	4
ing cloud that is to say both mixed-phase	410 In	4
ur study, we find for a lower-level altitude	411 cl	4
ci et al., 2019), a potential radiative impact	412 cu	4
respectively. We now separate the Eastern	413 N	4
tica (Figure 5 in Listowski et al., 2019) for	414 ele	4
that Net $\Delta SR_{global}^{max} = 0.5-5.0$ W m <sup>-2</sup> in	415 th	4
<sup>2</sup> for the remaining seasons. Over Western	416 De	4
ore intense because of higher temperatures	417 A	4
ctic Plateau: Net $\Delta SR_{global}^{max} = 12.5-30.0 \text{ W}$	418 an	4
; 7.5-20.0 W m <sup>-2</sup> in March-April-May; 2.5-	419 m	4
<sup>2</sup> in September-October-November.	420 10	4

421

#### 422 6. Conclusions

423 Combining the observations of temperature, water vapour and liquid water path from a ground-based microwave radiometer, backscattering and depolarization from a ground-based 424 425 LIDAR and surface radiations at long and short wavelengths, our analysis has been able to 426 evaluate the presence of supercooled liquid water clouds over the Dome C station in summer. 427 Focussing on 4 Decembers (2018-2021), the sensitivity of the SLWCs to temperature and LWP 428 has been established with temperature logarithmically increasing from -36.0°C to -16.0°C when liquid water path increases from 1.0 to 14.0 g m<sup>-2</sup>. We have also evaluated that SLWCs 429 positively affect the net surface radiation, which logarithmically increases from 0.0 to 50.0 W 430 431 m<sup>-2</sup> when liquid water path increases from 1.7 to 3.0 g m<sup>-2</sup>. Our study clearly shows that: 1) SLWCs have a positive impact on LWD and LWU, with  $\Delta SR$  increasing from 0 to 100 W m<sup>-2</sup> 432 and from 0 to 40 W m<sup>-2</sup> for LWP ranging from 1.0 to 4.0 and from 1.6 to 2.5 g m<sup>-2</sup>, respectively, 433





- and 2) SLWCs have a clear negative impact on SWD and SWU, with  $\Delta SR$  decreasing from 0 to -140 W m<sup>-2</sup> and from 0 to -75 W m<sup>-2</sup> with LWP ranging from 1.2 to 3.8 and from 1.2 to 3.2 g m<sup>-2</sup>, respectively.
- 437 Finally, extrapolating the radiative impact of the SLWCs from the Dome C station to the 438 Antarctic continent shows that SLWCs have a great potential radiative impact all over Antarctica whatever the season considered, up to 5.0 W m<sup>-2</sup> over the Eastern Antarctic Plateau 439 and up to 30 W m<sup>-2</sup> over the Antarctic Peninsula in DJF. This stresses the importance of 440 441 accurately modelling SLWCs when calculating the Earth energy budget to adequately forecast the Earth climate evolution, especially since the climate is rapidly changing in Antarctica, as 442 443 illustrated by the surface temperature record of -12°C recently observed in March 2022 at the 444 Concordia station and largely publicized worldwide (see e.g. https://www.latimes.com/world-445 nation/story/2022-03-19/antarctica-and-arctic-70-and-50-degrees-above-normal).

446

#### 447 Data availability

HAMSTRAD data are available at http://www.cnrm.meteo.fr/spip.php?article961&lang=en
(last access: 3 May 2022). The tropospheric depolarization LIDAR data are reachable at
http://lidarmax.altervista.org/lidar/home.php (last access: 3 May 2022). Radiosondes are
available at http://www.climantartide.it (last access: 3 May 2022). BSRN data can be obtained
from the ftp server (https://bsrn.awi.de/data/data-retrieval-via-ftp/) (last access: 3 May 2022).

453

#### 454 Author contribution

PR, MDG, and AL provided the observational data. PR developed the methodology. All the
co-authors participated in the data analysis and in the data interpretation. PR prepared the
manuscript with contributions from all co-authors.

458





### 459 Competing interests

- 460 The authors declare that they have no conflict of interest.
- 461

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473

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601	Tables
602	Table 1. Time-coincident data availability (green) in Decembers 2018-2021 for HAMSTRAD

603 temperature and LWP, Lidar Backscattering and Depolarization and BSRN Surface Radiances

604 (Net, LWD, LWU, SWD and SWU). The 5 clear-sky (Reference) days are highlighted in red.

Year		December Days																													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31
2018																															
2019																															
2020																															
2021																															





607

- 608 **Table 2.** Gaussian functions fitted to the N(x) function for  $x = \theta$  (°C) or  $\Delta SR$  (W m<sup>-2</sup>). Units
- 609 of  $a_1$ ,  $a_2$ ,  $a_3$ , and c are in count number for  $\theta$  and  $\Delta SR$ ; units of  $\mu_1$ ,  $\mu_2$ ,  $\mu_3$ ,  $\sigma_1$ ,  $\sigma_2$ , and  $\sigma_3$  are
- 610 in °C for  $\theta$  and in W m<sup>-2</sup> for  $\Delta SR$ .

x	<i>a</i> <sub>1</sub>	μ <sub>1</sub>	$\sigma_1$	<i>a</i> <sub>2</sub>	<b>μ</b> 2	$\sigma_2$	<i>a</i> <sub>3</sub>	μ3	$\sigma_3$	С
θ	15.0 10 <sup>3</sup>	-31.5	1.45	5.0 10 <sup>3</sup>	-28.0	1.65	0.5 10 <sup>3</sup>	-19.0	2.5	<b>-9</b> .1 10 <sup>-6</sup>
Net $\Delta SR$	2106.5	0.02	19.2	941.4	29.8	22.0	-	-	-	19.5
LWD $\triangle SR$	1010.8	80.1	21.9	1565.6	10.0	23.9	-	-	-	18.4
LWU ∆ <i>SR</i>	1476.4	-10.0	14.9	1834.7	25.0	16.2	-	-	-	185.4
SWD $\Delta SR$	1317.2	-5.0	15.8	717.4	-80.0	64.7	-	-	-	9.1
SWU ∆ <i>SR</i>	1928.8	-5.0	19.2	1163.4	-59.9	17.6	-	-	-	9.1

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Table 3. Coefficients of the relations  $f(LWP) = a + b \ln(LWP)$  for the temperature θ or surface radiation anomalies ΔSR. Units of θ and ΔSR, as well as of their corresponding "a" values are in °C and W m<sup>-2</sup>, respectively; units of b are in °C g<sup>-1</sup> m<sup>2</sup> for θ and in W / g for ΔSR; units of LWP are in g m<sup>-2</sup>. The last column shows the range of LWP values for which the relation is valid (in black), and in red (blue) the sub-range in which a positive (negative) impact is observed on ΔSR. Note that  $a \pm \delta a$  corresponds to the range of a values where the relationship is valid.

f(LWP)	$a \pm \delta a$	b	Valid range	Valid range				
			for $\boldsymbol{\theta}$ or $\Delta SR$	for LWP				
θ	$-33.8 \pm 1.5$	6.5	[-36; -16]	[1.0; 14.0]				
Net $\Delta SR$	$-50.0 \pm 10.0$	90.0	[-15; 50]	[1.5; 3.0] / <b>[1.7; 3.0]</b>				
LWD $\triangle SR$	$5.0 \pm 15.0$	65.0	[-10; 100]	[0.8; 4.0]/ <b>[1.0; 4.0]</b>				
LWU $\Delta SR$	$-45.0 \pm 30.0$	90.0	[-20; 40]	[1.3; 2.5]/ <b>[1.6; 2.5]</b>				
SWD $\Delta SR$	$30.0 \pm 30.0$	-130.0	[-140; 10]	[1.1; 3.8]/ [1.2; 3.8]				
SWU $\Delta SR$	$15.0 \pm 15.0$	-75.0	[-75;10]	[1.1; 3.2]/ [1.2; 3.2]				

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Figure 1: (From top to bottom): Time evolution (UTC, hour) of the Lidar Backscattering Signal, the Lidar Depolarization Signal, the HAMSTRAD LWP and the HAMSTRAD temperature profile measured on 27 December 2021. The time evolution of the SLW cloud (as diagnosed by a backscattering signal > 60 A.U. and a depolarization signal < 5%) is highlighted by the red and grey areas in the third and the forth panel from the top, respectively. The height above the ground is shown on the third panel from the top with the y-axis on the right. The 00:00 and 12:00 local times (LT) are highlighted by 2 vertical dashed lines.







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634 Figure 2: (Top) Time evolution (UTC, hour) of the SLWC (red areas) on 27 December 2021.

635 (Bottom, from left to right) Snapshots from the HALO-CAM video camera taken on: 01:00 (no

636 SLWC), 07:19 (SLWC), 09:00 (no SLWC), 10:14 (SLWC), 13:00 (no SLWC), 16:03 (SLWC),

- 637 18:01 (no SLWC) and 20:53 UTC (SLWC). The 00:00 and 12:00 local times (LT) are
- 638 highlighted by 2 vertical dashed lines.

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Figure 3: Time evolution (UTC, hour) of the clear-sky surface radiations (SR, W m<sup>-2</sup>) observed
by the BSRN instruments on 2 December 2018 (red), 19 December 2018 (orange), 3 December
2021 (green), 17 December 2021 (blue) and 26 December 2021 (black): (from top to bottom)
Net SR, Longwave Downward SR (LWD SR), Longwave Upward SR (LWU SR), Shortwave
Downward SR (SWD SR) and Shortwave Upward SR (SWU SR). The 00:00 and 12:00 local
times (LT) are highlighted by 2 vertical dashed lines.







**Figure 4:** Time evolution (UTC, hour) of the clear-sky surface radiation variability ( $\delta$ SR<sub>Ref</sub>, W m<sup>-2</sup>), namely the clear-sky surface radiations observed by the BSRN instruments on 2 December 2018 (red), 19 December 2018 (orange), 3 December 2021 (green), 17 December 2021 (blue) and 26 December 2021 (black) minus the corresponding values averaged over the 5 cloud-free days: (from top to bottom) Net SR, Longwave Downward SR (LWD SR), Longwave Upward





- 654 SR (LWU SR), Shortwave Downward SR (SWD SR) and Shortwave Upward SR (SWU SR).
- The 00:00 and 12:00 local times (LT) are highlighted by 2 vertical dashed lines.

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**Figure 5:** Time evolution (UTC, hour) of the Surface Radiation Anomaly ( $\Delta SR$ ), difference between the SR (W m<sup>-2</sup>) measured on 27 December 2021 and the reference clear-sky ( $SR_{Ref}$ ) SR (W m<sup>-2</sup>) measured on 26 December 2021: (from top to bottom) Net (Net  $\Delta SR$ ), longwave downward (LWD  $\Delta SR$ ), longwave upward (LWU  $\Delta SR$ ), shortwave downward (SWD  $\Delta SR$ ) and shortwave upward (SWU  $\Delta SR$ ). The time evolution of the SLW cloud is highlighted by a red area in the uppermost panel, with the height on the y-axis shown on the right. The 00:00 and 12:00 local times (LT) are highlighted by 2 vertical blue dashed lines.







667 Figure 6: (Left) Probability Density Function (PDF, %) of the Temperature (°C) as a function of Liquid Water Path (LWP, g m<sup>-2</sup>) contained in the Supercooled Liquid Water clouds (SLWCs) 668 above Dome C in December 2018-2021. The PDF is defined in the text. (Centre) Weighted-669 670 average LWP vs. temperature with a fitted logarithmic function (blue solid) encompassing the 671 significant points (2 dashed blue lines). Horizontal bars represent 1-sigma variability in LWP 672 per 1°C-wide bin over significant points. (Right) Temperature as a function of count number 673 per 1°C-wide bin (black solid line) with 3 fitted Gaussian functions (red dashed curves). The 674 sum of the 3 Gaussian functions is represented by a red solid line.

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Figure 7: (Left) Probability Density Function (PDF, %) of the Net Surface Radiation Anomaly 678 (Net  $\Delta SR$ , W m<sup>-2</sup>) as a function of Liquid Water Path (LWP, g m<sup>-2</sup>) contained in the Supercooled 679 680 Liquid Water clouds (SLWCs) above Dome C in December 2018-2021. The PDF is defined in 681 the text. (Centre) Weighted-average LWP vs. Net  $\Delta SR$  with a fitted logarithmic function (blue 682 solid) encompassing the significant points (2 dashed blue lines). Horizontal bars represent 1sigma variability in LWP per 5 W m<sup>-2</sup>-wide bin over significant points. (Right) Net  $\Delta SR$  as a 683 function of count number per 5 W m<sup>-2</sup>-wide bin (black solid line) with 2 fitted Gaussian 684 685 functions (red dashed curves). The sum of the 2 Gaussian functions is represented by a red solid 686 line.







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689 Figure 8: As in Figure 7 but for the Longwave Downward (top) and Upward (bottom) Surface

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<sup>690</sup> Radiation Anomaly (LWD and LWU  $\Delta SR$ , respectively).







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694 Figure 9: As in Figure 7 but for the Shortwave Downward (top) and Upward (bottom) Surface

695 Radiation Anomaly (SWD and SWU  $\Delta SR$ , respectively).