



# Iberulite fall and formation mechanism during a Sahara dust event in Switzerland in February 2021

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**Abstract.** Sahara dust events can promote the formation of micrometer-sized spherical mineral aggregates known as iberulites as they are usually found on the Iberian Peninsula. Iberulites have not, to our knowledge, been reported from Central Europe. Two formation mechanisms – in- and below scavenging – have been given. Strong iberulite fall (IFs) were observed in Western Switzerland in February 2021 when large amounts of dust were transported from the Moroccan-Algerian border towards Central Europe during a Saharan Dust Event (SDE). In contrast to the previous IFs observed on the Iberian Peninsula, this IF occurred under low-temperature conditions e.g. near the freezing point in the cloud and at the surface. The relative humidity in the first 1000 m below the clouds decreased from 70 to 40%. and the comparison between particle-size distribution (PSD) within the iberulites and the dust PSD in the free atmosphere revealed a Greenfield gap in the iberulites, e.g., a lack of particles in the range 0.1-0.3µm relative to the dust in the free atmosphere. The meteorological conditions, the microstructure of the iberulites, and the presence of a Greenfield gap point to below-cloud scavenging as the most likely formation mechanism. Further discoveries of iberulite in Swiss SDE samples suggest that the phenomenon is widespread and not only limited to regions close to the dust source.





## 30 1 Introduction

31 Sahara Dust Events (SDEs) reach about 10-40 times per year the European Alps and significantly contribute to the aerosol  
32 load above the region, with strongest influence from February to June, and October to November (Coen et al., 2004; Collaud  
33 Coen et al., 2025; Mauro et al., 2019; Flentje et al., 2015). SDEs in February and March particularly increased since 2020  
34 (Cuevas-Agulló et al., 2024). Aerosol concentrations during these events often exceed the European and Swiss air quality  
35 standards and the WHO PM<sub>10</sub> air quality guidelines. There is evidence that desert dust affects human health, e.g., increases  
36 in cardiovascular mortality and respiratory morbidity, but the evidence remains inconsistent when considering the dust's  
37 geographical origin (Fussell and Kelly, 2021; Tobias et al., 2019). Dust also affects climate and ecosystems (Buseck and  
38 Pósfai, 1999; Parajuli et al., 2022). Although the importance of dust is recognized, dust mineralogical studies are scarce  
39 (Engelbrecht and Derbyshire, 2010).

40 A special form of dust aerosol particles are giant spherule-shaped aggregates between 50 and 200  $\mu\text{m}$  in diameter. These  
41 “microspherulites” are called iberulites, in reference to their first observation in southern Spain (Díaz-Hernández and Párraga,  
42 2008). Such iberulites were also reported from Mallorca (Spain) (Fiol et al., 2005) and from Tenerife (Spain) (Cuadros et al.,  
43 2015), but no observations have been reported from regions North of the Alps. Such aggregates are thought to form inside  
44 raindrops by the coalescence of smaller droplets in a cloud, thereby increasing the number of condensation nuclei in the latter,  
45 or by the capture of particles below a cloud by drops as they fall to the ground (Díaz-Hernández and Párraga, 2008). A major  
46 SDE hit Switzerland at the beginning of February 2021. Systematic SDE monitoring is conducted in Switzerland at the  
47 Jungfraujoch Sphinx observatory (inlet altitude 3585 m above sea level ASL (Vollmer et al., 2020)) since 2001 (Collaud Coen  
48 et al., 2025). Due to its elevation and the negligible local emissions, Jungfraujoch is a suitable location to investigate long-  
49 range transport processes (Herrmann et al., 2015). The event on February 6<sup>th</sup> was detected as one of the strongest SDEs since  
50 its systematic monitoring in 2001 (Collaud Coen et al., 2025). The dust reached the Alps on February 6<sup>th</sup>, 2021 in the afternoon.  
51 At Jungfraujoch, a record PM<sub>10</sub> concentration of more than 700  $\mu\text{g m}^{-3}$  was measured (NABEL, BAFU/Empa). The IF  
52 analyzed here happened at lower altitudes in the village of Saint L  gier (canton of Vaud, 46.471   N, 6.877  E), the city of  
53 Fribourg (canton of Fribourg, 46.717   N 7.083   E), and the city of Geneva (canton of Geneva, 46.033   N, 6.117   E), all in  
54 western Switzerland. Until now, two formation processes of iberulites have been proposed: in- and below-cloud scavenging.  
55 The goal of the present work is to explore the aggregation mechanism in greater detail, leveraging the well-documented  
56 meteorological conditions during the fall in our case.



## 57 2 Methods

### 58 2.1 Collection sites

59 The fall of the Iberulites was observed on the early afternoon of February 6th in St. L  gier, Fribourg, and Geneva  
60 (Switzerland). The city of Fribourg (500-700 m ASL) is 50 km NNE of Lake Geneva on the Swiss plateau, and the village of  
61 St. L  gier (576 m ASL) is on the central northern shore of the same lake. The geology of the surroundings of both sites is  
62 dominated by calcareous mountains (Jura, Pr  alpes) (Tr  mpy, 1980; Matzenauer, 2011). In St.L  gier, the iberulites were  
63 picked up from the ground with tweezers, and in Fribourg, an aluminum foil was laid on the ground, from which samples were  
64 collected with a fine spatula. The iberulites were gently deposited onto two different substrates: head specimen mounts and  
65 boron substrates (see below).

66 The high-Alpine station Jungfraujoch (7.98   N, 46.55   E; 3580 m ASL), lies in the northern Swiss Alps. Its remote location,  
67 far from local anthropogenic sources, makes it ideally suited for monitoring background atmospheric conditions over central  
68 Europe. Jungfraujoch hosts a comprehensive long-term in-situ monitoring program for aerosols and trace gas as a contribution  
69 to the Global Atmosphere Watch (GAW) program of the World Meteorological Organization (WMO) (Bukowiecki et al.,  
70 2016) and is part of the Swiss National Air Pollution Monitoring Network (H  glin, 2021).

### 71 2.1 Meteorological data

72 Meteorological data were obtained from radio sounding balloons, which are launched by the Federal Office for Meteorology  
73 and Climatology of Switzerland (MeteoSwiss) station in Payerne (46.817   N, 6.933   E ), twice a day (23:00 and 11:00 h UTC).  
74 Payerne is located 17 km west and 47 km north of the collection sites in Fribourg and St. L  gier, respectively. The station is  
75 equipped with a Raman lidar, ceilometers, and a Doppler radar to measure precipitation, aerosol concentration, and wind  
76 profiles. On February 6th, the ceilometer profile series from Payerne exhibited gaps in the morning and smaller ones in the  
77 afternoon due to ground fog. The area is also covered by two weather radars: one on La Dole Mountain (46.425  N, 6.099  E ),  
78 west of St. L  gier and near Geneva, and the other on the Plaine Morte (46.383  N, 7.500  E ), in the central Bernese Alps  
79 southeast of the two collection sites.

80 The size distributions of fine-mode and coarse-mode aerosol at JFJ have been measured by a home-built Scanning Mobility  
81 Particle Size Spectrometer (SMPS) (Wiedensohler et al., 2012; Jur  nyi et al., 2011) and by a white-light optical particle size  
82 spectrometer (FIDAS, Palas GmbH, Germany) (Fischer and H  glin, 2024), respectively.

83 The dust RGB product, based on three infrared channels of the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) on  
84 EUMETSAT's MSG-11 satellite (<https://www.eumetsat.int/0-degree-service>), was used to determine the origin of the dust  
85 arriving in the afternoon over western Switzerland. The plume trajectory was traced on satellite images using the approach  
86 described by (Ashpole and Washington, 2013). The visually determined dust trajectories were verified by both forward and  
87 backward trajectory calculations using the Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model



88 developed by the National Oceanic and Atmospheric Administration's (NOAA) Air Resources Laboratory (ARL) (Draxler and  
89 Rolph, 2003). HYSPLIT uses archived 3-dimensional meteorological fields generated from ground and satellite observations,  
90 as well as short-term forecasts. For the present back-trajectory calculations, the Global Forecast System (GFS) dataset from  
91 the National Centers for Environmental Prediction (NCEP) at 0.25° resolution was used. The backward trajectories were  
92 calculated for an arrival time over Fribourg at an altitude of 2000 m above ground level (AGL) between 12:00 and 18:00 UTC  
93 on February 6th (approximately the level of the highest density of the arriving cloud). The forward calculations were done  
94 with a starting point in the center of the first dust plume observed in the satellite images.  
95 The source meteorological data were extracted using the Real-time Environmental Applications and Display sYstem (READY)  
96 from the archived Global Data Assimilation System (GDAS) (Rolph et al., 2017). The data were obtained for the center of the  
97 primary dust source observed in the satellite image, e.g., 33.8°N, 1.5°W.

## 98 **2.3 Scanning Electron (SEM) and Optical Polarization Microscopy (OPM) analyses**

99 Single particles and iberulites were separated from bulk samples using an Olympus SZX12 binocular (Tokyo, Japan). They  
100 were transferred with a fine spatula and gently deposited onto a ½" pin stub mount (Ted Pella, Redding, CA, USA) previously  
101 coated with 12 mm Pelco tabs (Ted Pella, Redding, CA, USA) or onto homemade polished boron substrates. The latter were  
102 used to improve the contrast of low-atomic-number particles, such as soot (carbon). Each sample (area depending on particle  
103 load on the sample surface) was analyzed using automated SEM/EDX single-particle analysis (also known as Computer-  
104 Controlled SEM, CCSEM). For this purpose, a Zeiss Gemini 300 Field Emission Gun (FEG)-SEM equipped with an Oxford  
105 X-MAX EDS detector with an 80 mm<sup>2</sup> window, a high-efficiency four-quadrant Back Scattered Electron (BSE) detector, and  
106 the particle analysis software included in AZtecFeature (Oxford Instruments) at Particle Vision (Fribourg, Switzerland) was  
107 used. The analyses were performed at 20 kV. Particles deposited on the boron substrate were recognized by simple gray-value  
108 (i.e., BSE intensity) thresholding. EDX analyses were performed for each recognized particle. A "ZAF" correction (PAP  
109 version of the  $f(\rho z)$  formalism (Pouchou and Pichoir, 1985) was applied automatically by the AZtec software to the raw data  
110 to calculate elemental concentrations. Quantification of the analyses was performed using factory-delivered standard  
111 measurements. The ZAF procedure is valid only for homogeneous samples with smooth surfaces perpendicular to the beam  
112 and considerably thicker than the excitation volume. Neither condition is met for the particles in our case. However, several  
113 authors (Weinbruch et al., 1997; Meier et al., 2018; Kandler et al., 2018) have shown that for particles larger than 1 µm in  
114 diameter, the relative standard deviation of corrected concentrations of major elements is approximately 2%. In contrast, for  
115 minor elements, the deviation is considerably larger (Kandler et al., 2018). As only particles with diameters > 1 µm were  
116 considered in the further processing of the data, the ZAF-corrected concentration values were retained without applying more  
117 sophisticated correction procedures. The standard deviation in ZAF-corrected concentrations measured in particles increases  
118 proportionally with the acceleration voltage (equivalent to increasing excitation volume). The measurement voltage (12 kV)  
119 is the lowest possible to excite characteristic X-ray lines for all elements present in the sample. Several blank analyses of empty



boron substrates have shown that under the applied analytical conditions, the substrate surface contains only the element boron and that in an area of ca. 1 mm<sup>2</sup>, a maximum of five particles were present on unexposed substrates. These particles are organic in nature and most likely follicles. Images obtained with both the optical microscope and the SEM were used to characterize the size distribution of the iberulites and their microstructure using Avizo software (Thermo Fisher). The picked iberulites were placed in a round 1-inch container and filled with an impregnation resin. The resulting cylindrical samples were polished on the top side containing the iberulites and analyzed with a FEI Sirion FEG microscope (20 kV) equipped with the same EDS system as the Zeiss microscope above. A BSE image of the polished section was also segmented using Avizo software (Thermo Fisher), allowing the counting of individual particles and the determination of their equivalent spherical diameter, as well as the corresponding size distribution. Due to the resolution of the segmentation process, only particles larger than 0.05 µm are possible to be recognized and counted.

## 2.4 X-ray diffraction

A loose single dust particle sample, containing no iberulites but collected during the same period when the iberulites fell in Fribourg, was analyzed with a Rigaku Ultima IV diffractometer equipped with a Position Sensitive Detector (PSD). Diffractograms were recorded from 5° to 70° 2θ in step-scan mode (0.02°/step and 1 min/°). The copper X-ray tube was operated at 40 kV and 40 mA. The CuKα-radiation was filtered with a nickel foil. Quantitative phase analysis by Rietveld refinement (Bish and Post, 1993) was performed on the recorded patterns using the PDXL2 software package (Rigaku). The refined instrumental parameters are the zero shift, the sample displacement, and the peak shape, modelled using a Pseudo-Voigt profile. The structure parameters for the phases present in the sample were taken from the International Centre for Diffraction Data database, but only the scale factor, the unit-cell parameters, the isotropic overall temperature factor, and, in some cases, the preferred-orientation parameter (phyllosilicates, calcite) were refined using the March-Dollase (Dollase, 1986) formalism. The fractions of the phases present were determined through the scale factors. The atomic positions and site occupancies were held fixed during refinement. Iterations were performed until the refinement converged.

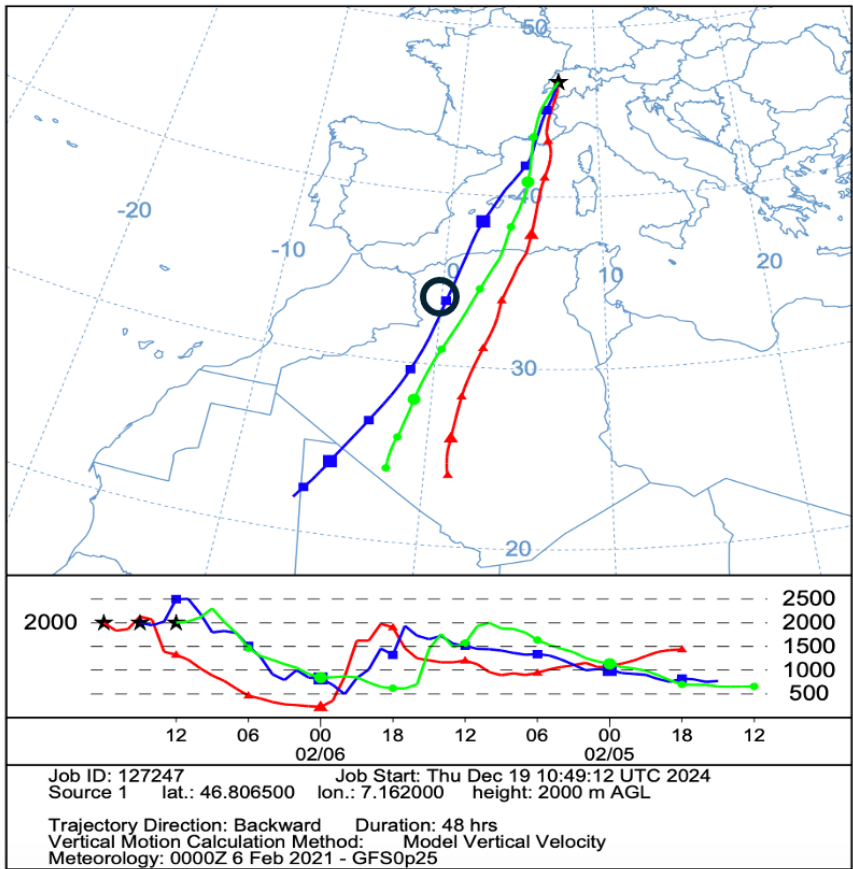
## 3 Results

### 3.1 Dust source and trajectory

The HYSPLIT backtrajectory calculations for air masses arriving over Fribourg in the early afternoon of February 6th originated in NW Africa (Fig. 1) between the southern Moroccan border and northwestern Algeria, a region. Satellite images showed the appearance of a dust plume (Fig. 2) in a region called "Haut Plateau". The air masses arriving during the afternoon and evening of February 5th over the Haut Plateau are at altitudes between 1000 m and 2000 m AGL, i.e., well within the Planetary Boundary Layer (PBL, Fig. 3). On February 5th, the upper limit of the PBL in the Haut Plateau region moved up to

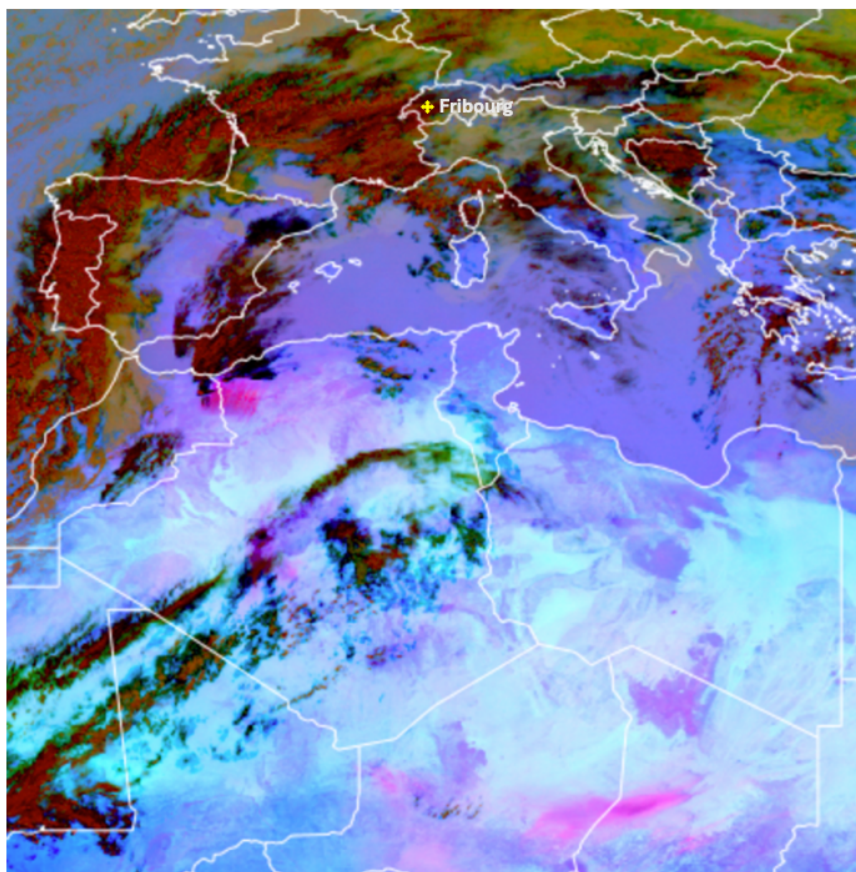


149 3500 m AGL, and the velocity in the surface air layer (10 m above ground) increased from 40 km/h to about 70 km/h. Weather  
150 stations in the Atlas Mountains recorded hurricane-force gusts (e.g., 117 km/h on 05.02.21 in Midelt, Morocco (Hoshyaripour,  
151 2021). This is clearly above the threshold wind speed for dust emissions given for western Algeria (Marticorena et al., 1997).  
152



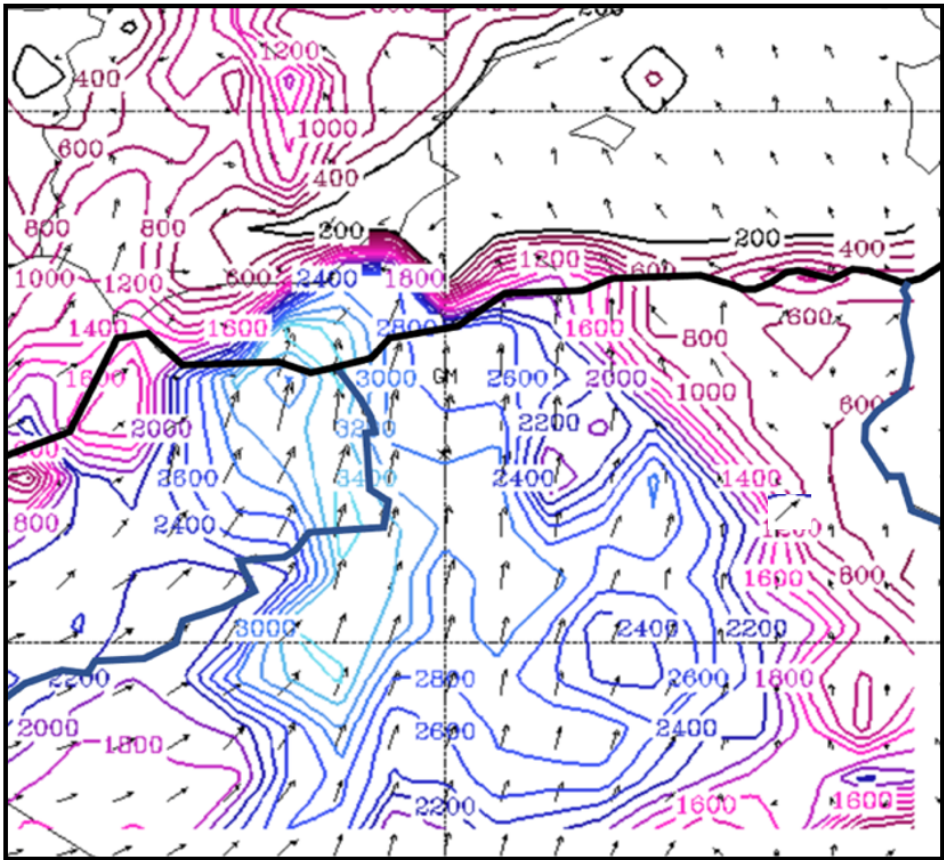
**Figure 1:** Back trajectories for air masses arriving over Payerne at an altitude of 2000m AGL during February 6th between 12:00 and 18:00 UTC. The black circle shows the location of the first appearance of the dust plume entrained across the Mediterranean Sea towards central Europe .





**Figure 2:** Satellite RGB image (5.2.2021. 22:00 UTC) from the SEVIRI dust product, showing the presence of dust by pink colors. The dust plume transported to Central Europe is seen east of the Moroccan – Algerian border.

153  
154  
155



↑ Wind vectors (knts) at 10m AGL  
|| Planetary Boundary Layer Height (m)

**Figure 3:** Map showing the upper boundary in meters AGL of the PBL over the Maghreb on February 5th at 12 h UTC. The arrows are wind vectors on the ground. The latter increases with increasing PBL depth (READY output (Rolph et al., 2017)).

156

157 The dust plume was entrained in a NE direction toward the Mediterranean Sea, where a low-pressure system formed. The  
158 associated warm conveyor belt lifted the dust to higher altitudes and transported it with the polar jet across the Mediterranean  
159 Sea toward the Alps, a process referred to as an "atmospheric river" (Francis et al., 2022; Rautela et al., 2024). Source  
160 sensitivities were calculated for Saharan dust (fine mode) arriving at JFJ using FLEXPART to obtain the backward trajectories  
161 and to detect general surface contact of air masses over the Saharan desert (Collaud Coen et al., 2025). The calculated  
162 sensitivities indicate that the direct pathway from the Sahara over the Mediterranean to the Alps is the most frequent.

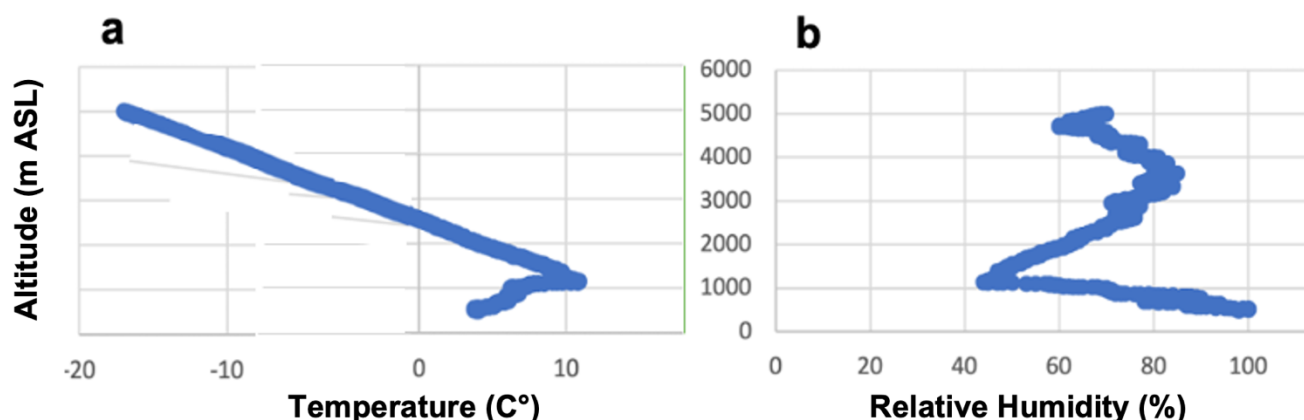
### 163 3.2 Local meteorology

164 Ground temperature measurements and the emagram from the radio sonde released at 11:00 UTC in Payerne revealed relatively  
165 high temperatures for February 6th in Western Switzerland (8° C at the ground and a zero-degree level at 2500 m ASL) and a



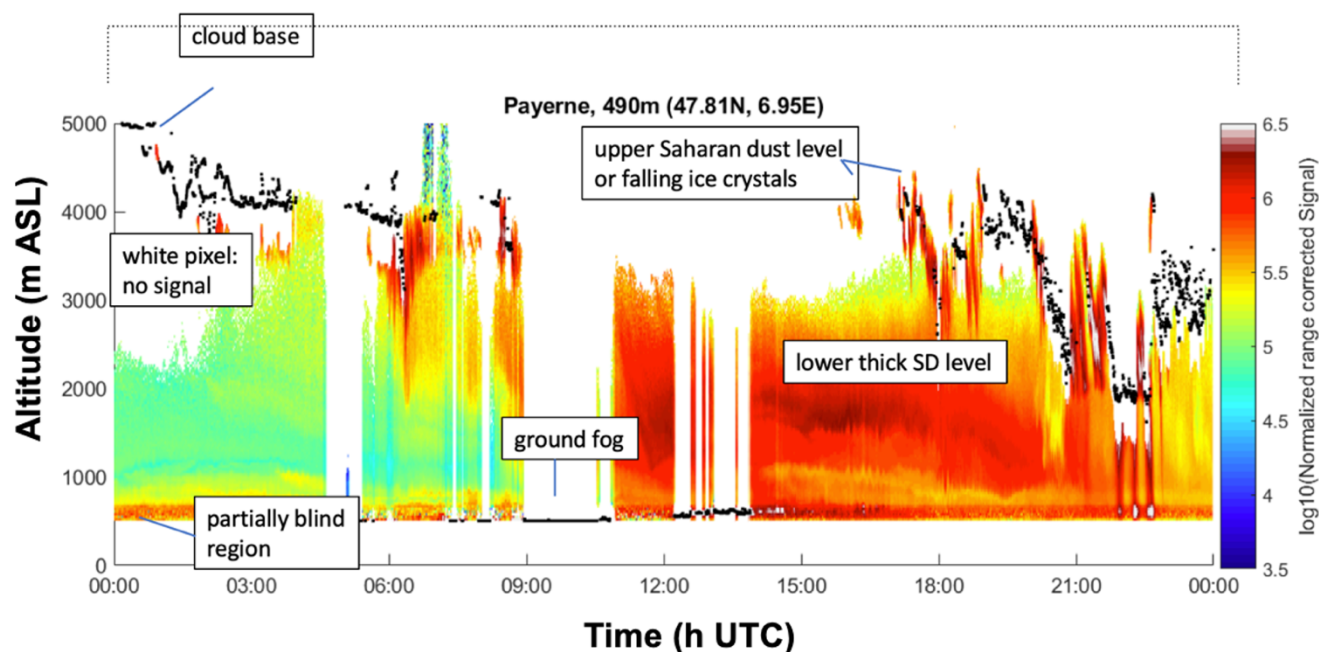


166 typical inversion situation (Fig. 4a). In contrast to conditions during previous IFs over the Iberian Peninsula (Diaz-Hernandez  
 167 and Sanchez-Navas, 2016) the temperature was much lower. The vertical profile of relative humidity (RH) was complex. RH  
 168 decreased from 100 % at the ground level to a very low 40 % at 1000 m, then increased to 85 % at 3500 m ASL (Fig. 4b).



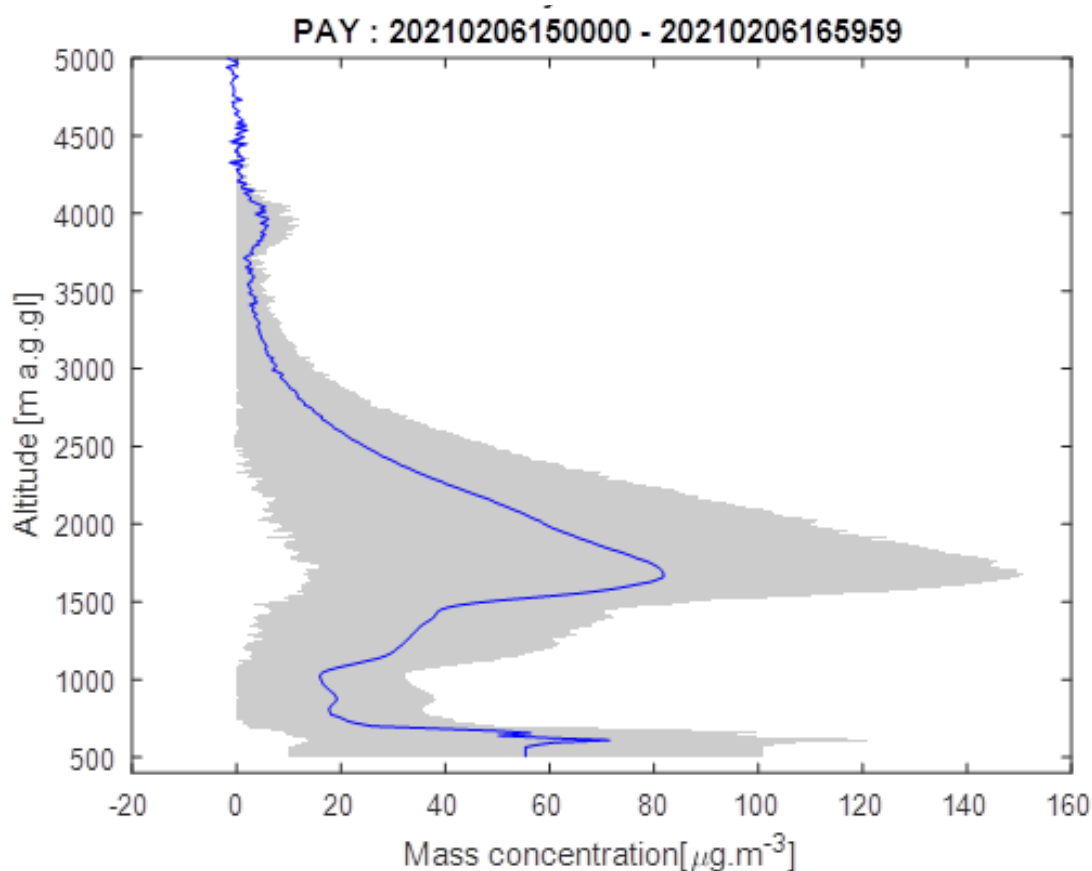
169  
 170 **Figure 4:** Temperature (a) and relative humidity (b) as a function of altitude from the radio sounding. The sounding balloon was released in  
 171 Payerne at 11:00 h UTC (MeteoSwiss).

172  
 173 The Payerne ceilometer time profile (Fig. 5) shows a cloud base at 5000 m ASL at midnight, with reflections that may be due  
 174 to aerosol (SDE) or ice particles, extending from the cloud base down to 3500 m. Early in the morning, the cloud base lowered  
 175 to 4000 m, and the reflections extended down to 2500 m. The below-cloud reflections have a streaky appearance in the  
 176 ceilogram. After a large gap in ceilometer recordings between 9:00 and 11:00 h UTC, a reflection wall extending from 3500  
 177 m to the ground appeared. These strong reflections from dust particles persisted throughout the day. The cloud base lowered  
 178 from 4000 m to 2000 m during the evening. At around 22:30 h, three short periods with very strong reflections were observed;  
 179 during those times, precipitation was recorded on the ground. After 23:00 h, the reflections became less intense.



**Figure 5:** Ceilogram profile for February 6<sup>th</sup> (MeteoSwiss)

Lidar measurements were used to derive a mass-concentration profile (Fig. 6) across the dust cloud, with the altitude range between 4000 and 5000 m serving as the "no aerosol" reference. The profile was derived using a lidar ratio of  $0.57 \text{ m}^2/\text{sr} \pm 9$  sr at 532 nm (Haarig et al., 2022), an Ångström coefficient of 0.5 for long-range transported dust in northern European regions (Ansmann et al., 2001), and a mass extinction ratio at 532 nm of  $0.47 \pm 0.04 \text{ m}^2 \text{ g}^{-1}$  (Nemuc et al., 2013). The profile exhibited three peaks. The maximum concentration of  $81 \mu\text{g m}^{-3}$  was observed at an altitude of 1675 m. The second-largest concentration was recorded just above the ground, and a third small peak was located just above the cloud base at approximately 3500 m.



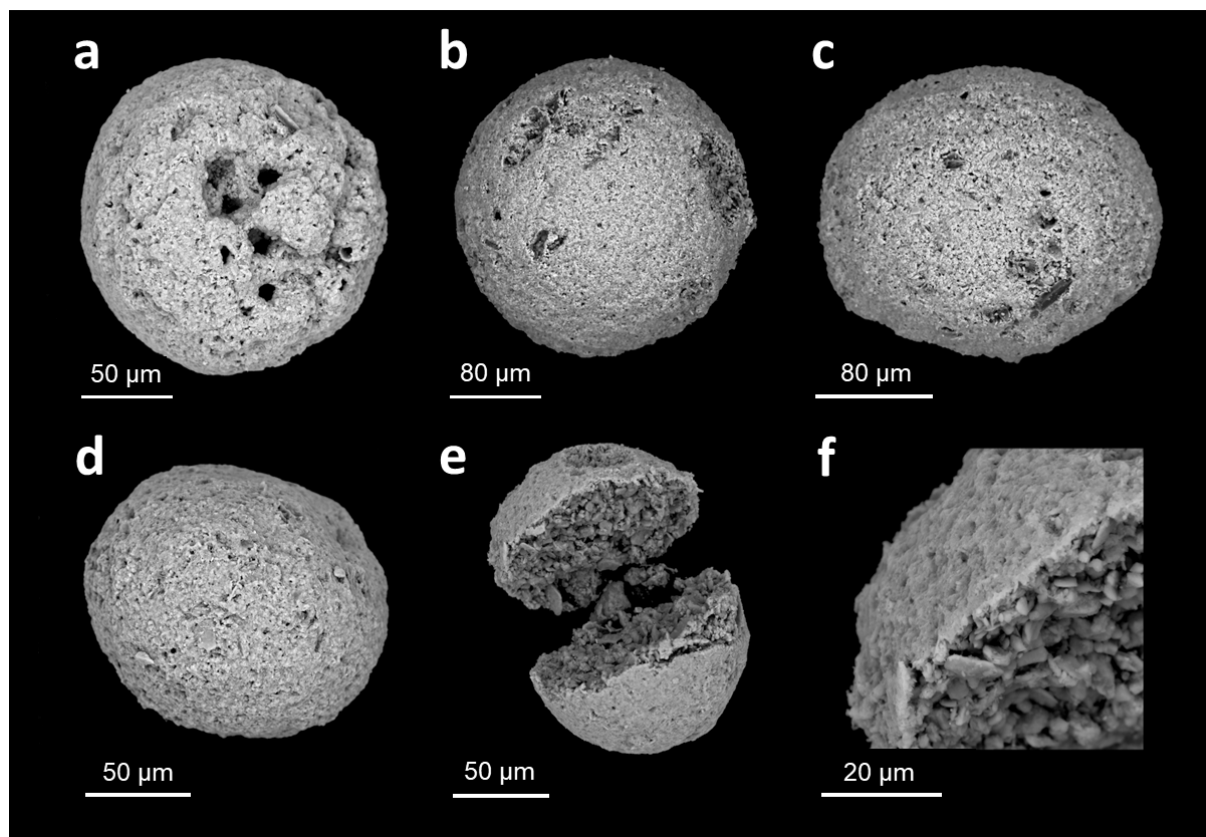
189  
 190 **Figure 6:** Aerosol mass concentration profile across the dust cloud (MeteoSwiss).

### 191 3.3 Iberulites collected in Fribourg and St. L  gier

192 In Fribourg, very large particles with diameters exceeding 30  $\mu\text{m}$  were detected. Unusual giant single particles have been  
 193 observed previously following long-range transport of Saharan dust (Van Der Does et al., 2018). Rapid transport, high  
 194 turbulence, and electrical forces are thought to compensate for the particles' weight. In our case, the large "particles" were not  
 195 single particles but multimineral particle aggregates with spherical to slightly ellipsoidal shapes, known as iberulites after their  
 196 first recording on the Iberian Peninsula (Fig. 7) (D  az-Hern  ndez and P  rraga, 2008). The cores of the iberulites are composed  
 197 mainly of coarser mineral grains (Fig. 8 e,f, and Fig. 9) that have accreted without a visible cement matrix. Except for a thin,  
 198 dense surface layer, the iberulite cores show no internal structure, such as layering or preferential grain orientation. The  
 199 iberulites collected in St L  gier ( $n = 425$ ) are larger than the Iberian specimens, with a mean diameter of 113  $\mu\text{m}$  (range: 43 –  
 200 365  $\mu\text{m}$ ).

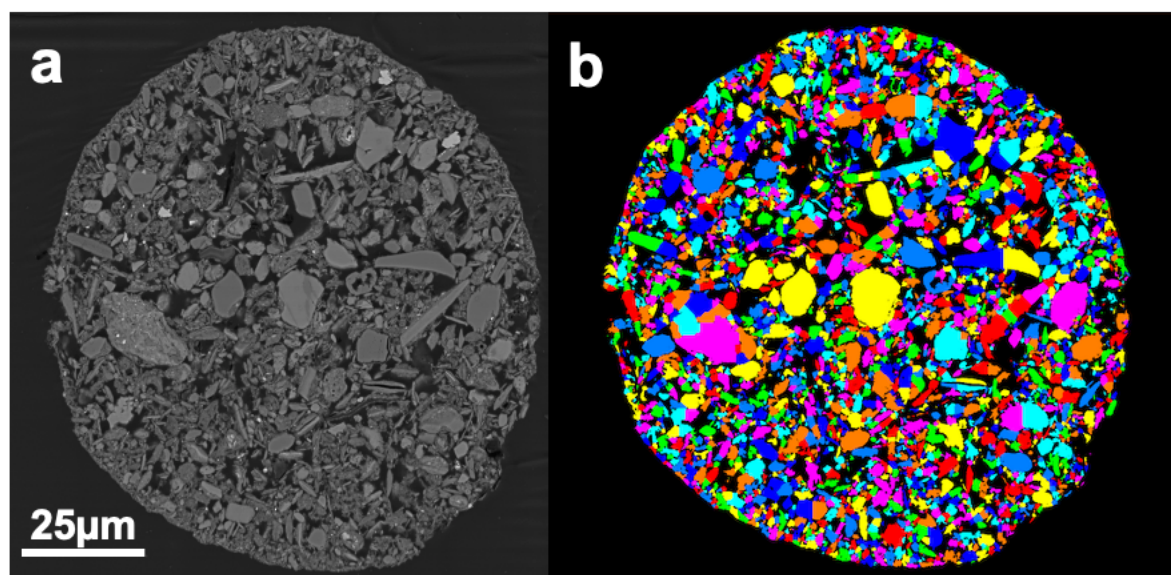


**Figure 7:** Light microscopy image of iberulites collected in St Léger.



**Figure 8:** SEM images of some iberulites collected in St L  gier on February 6th, 2021. Figure 8 f shows the fine-grained rim of an iberulite.





**Figure 9:** (a) BSE image of a polished section through an iberulite, and (b) the corresponding segmentation obtained from AVIZO.

Most iberulites have smooth surfaces with few large surface pores (Fig. 8), most likely due to particles that have been ejected from the aggregates. Spherical surface depressions, e.g., vortices formed by the flow of air around the aggregates (Díaz-Hernández and Párraga, 2008), are rare.

### 3.4 PSDs at JFJ and inside the iberulites

The long-term PSD measured with SMPS and CPC at JFJ, when the latter was in the cloud, was taken as representative of the PSD in the main dust layer. During times when JFJ is cloud-free, above the PBL, and no Saharan dust present the PSD has two modes: a stronger one near  $0.05\ \mu\text{m}$  and a weaker one between  $0.15$  and  $0.4\ \mu\text{m}$  (Nessler et al., 2003; Herrmann et al., 2015; Bukowiecki et al., 2016). The PSD measured at 11:00 UTC on February 6th during the SDE is monomodal, with a maximum near  $0.150\ \mu\text{m}$  (see Fig. 10, yellow bars), and shows much higher concentrations than measurements without SDEs, similar to PSDs observed in previous Saharan dust episodes at JFJ (Chou et al., 2011). Coarse-mode particles arriving at JFJ were well documented in previous particle volume distributions, with a mode at  $1.4\ \mu\text{m}$  (Schwikowski et al., 1995). Saharan dust clouds transported over long distances, such as to Barbados during the SALTRACE campaign (Weinzierl et al., 2017), to Aeronet sites in the eastern Caribbean (Velasco-Merino et al., 2018), over the Mediterranean Sea during the ChArMEx/ADRI-MED campaign (Denjean et al., 2016), and over Cape Verde during the SAMUN2 campaign (Weinzierl et al., 2011) have similar PSDs. The largest particles observed at the Caribbean sites are well above the maximum diameter expected for settling due to Stokes' law during transport. The modes observed at JFJ and in the campaigns cited above are



similar to the PSDs observed in the source regions (Panta et al., 2023; Bencherif et al., 2022; Dubovik et al., 2006), but are lower in concentration and lack particles  $> 30\mu\text{m}$ .

The equivalent volume diameter  $d_{ve}$  and the shape factor  $\chi$  of the particles inside the iberulites were estimated following (Ott and Peters, 2008):

$$d_{ve} = \sqrt{\frac{4A}{\pi}} \quad (1)$$

$$\chi = \frac{p^2}{4\pi A} = \frac{1}{\text{circularity}} \quad (2)$$

with  $A$  the area covered by and  $p$  the perimeter of the particles in the segmented BSE image. The PSDs inside the iberulites are  $\pm$  monomodal with a maximum at  $2.5\mu\text{m}$  (Fig. 10, blue bars), i.e., much larger than the value for the Saharan dust sampled in JFJ, i.e.,  $0.15\mu\text{m}$  (Fig. 10, yellow bars). The PSD of the particles inside the iberulites shows a minimum where the JFJ has a maximum. The average aspect ratio (AR) compares to the median AR of long-range transported Saharan dust particles measured in the Caribbean during the PRIDE experiment over Puerto Rico (1.93 vs 1.9) (Reid et al., 2003), but is larger than the value obtained for short- to medium-range transported Saharan dust over Tenerife (1.64) (Kandler et al., 2007) and during the AMMA SOP0/DABEX campaign, e.g., samples collected on site and during flights over the western Sahara (1.7) (Chou et al., 2008). This increase in particle asphericity during transport is likely due to the preferential settling of spherical particles, which have a greater terminal fall speed than aspherical particles of the same volume (Mallios et al., 2020). In the present case AR ranges from 1.0 to 6.0 and, as previously reported (Huang et al., 2020), shows little dependence on dust size.

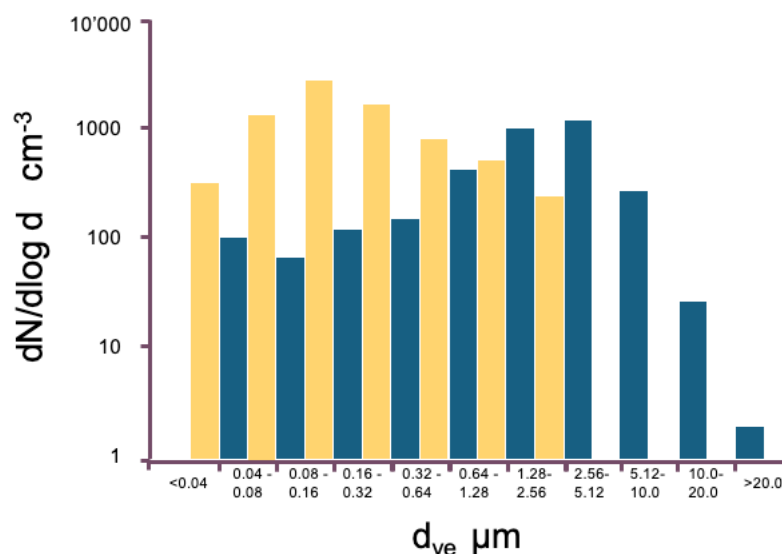
In order to compare the SMPS with the SEM derived PSDs,  $d_m$  of the first has to be converted to volume equivalent diameter  $d_{ve}$  (Decarlo et al., 2004):

$$d_{ve} = \frac{d_m C_c(d_m)}{C_c(d_{ve}) \chi} = CF d_m \quad (3)$$

with  $C_c(d_m)/C_c(d_{ve})$ , the Cunningham slip correction factor ratio between the mobility and volume equivalent diameters, the dynamic shape factor  $\chi$ , and the conversion factor  $CF$ .  $C_c(d_{ve})$  is unknown. The ratio  $C_c(d_m)/C_c(d_{ve})$  (the ratio given by Peng et al. is the reverse of the latter one!) has been measured only(?) for salt particles smaller than 600 nm. It is always less than one, increases with size, and decreases with aspect ratio (Peng and Liu, 2022). In contrast to the aspect ratios, shape factors appear to depend strongly on size. They are size-, flow-regime (Alexander et al., 2016), and orientation (Cheng et al., 1988) dependent, ranging from near unity for smaller particles to a median value of 3 for the largest particles. The average  $\chi$  of the particles inside the iberulites is 1.46, very close to 1.4 reported for most dust particles in the 2 to 10  $\mu\text{m}$  range (Kaaden et al., 2009). Overall,  $CF$  is mostly smaller than 1. The converted volume-equivalent diameters are smaller than the mobility diameters, and



the PSD shifts to the left. For a particle with a diameter of 0.2 or 2.0  $\mu\text{m}$ , both with an axis ratio of 1.93, and corresponding values of  $C_c(d_m)/C_c(d_{ve})$  and  $\chi$ , the shift is 1 bin to the left for both, e.g., the maximum of the JFJ PSD is shifted from the (0.16 – 0.32 $\mu\text{m}$ ) to the (0.16 – 0.32 $\mu\text{m}$ ) bin. Overall,  $d_{ve}$  is always smaller than  $d_m$ .

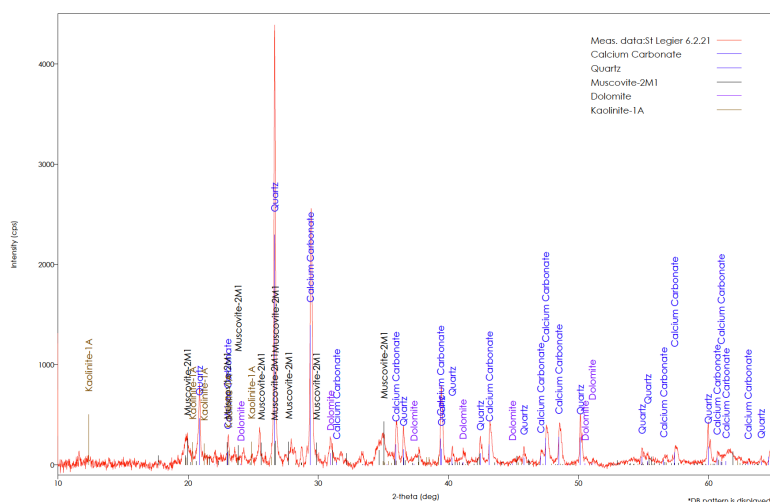


**Figure 10:** Yellow bars: Jungfraujoch PSD measured February 6th at 12:00 (shifted 1bin to the left relative to the original data, s. text), blue bars: particle size distribution measured for an iberulite sample collected in St. L  gier.

### 3.5 Composition of Saharan dust and iberulites

#### 3.5.1 XRD -results (Rietveld analysis) and single particle analyses

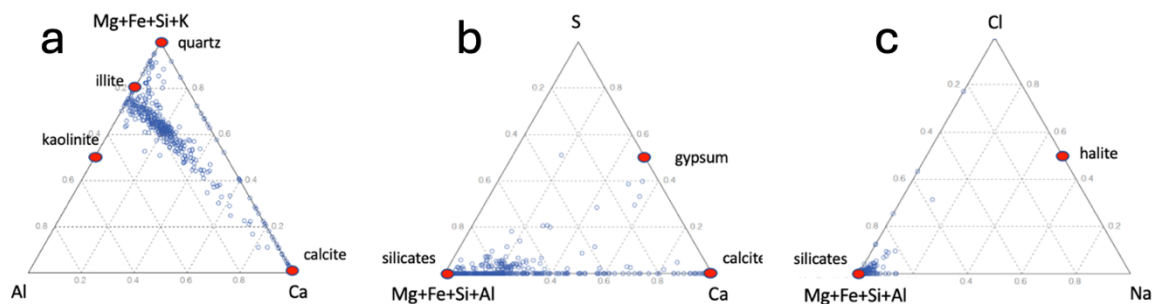
X-ray diffraction (Fig. 11) showed that iberulites contain phyllosilicates (illite, muscovite, and kaolinite, totaling 34.1 wt%), followed by carbonates (27.0 wt% calcite, 4.0 wt% dolomite) and quartz (23.3 wt%). The concentrations of gypsum and halite were below the detection limit.



**Figure 11** X-ray diffraction pattern of the St. Léger sample.

### 3.5.2 EDS analysis

Analysis of individual particles within an iberulite confirms the XRD results. EDS analyses on the polished iberulite section have a resolution of 1 – 2  $\mu\text{m}$ , depending on the thickness of the section and the local composition. Trends from the ideal muscovite composition toward the calcite composition, as well as toward the quartz compositions, in the (Mg+Fe+Si+K–Al–Ca) ternary diagram indicate that the analyses are mixtures of the three phases (Fig. 12 a). The ternary diagram (Mg+Fe+Si+K–S–Ca), with Mg+Fe+Si+K as the "silicate" corner and Ca as the "calcite" corner (Fig. 12 b), shows measurements mainly along the joint between the silicate and calcite corners, indicating mixed analyses between these corners, with some measurements scattered in the triangle silicates-calcite-gypsum, indicating mixed analyses among these phases or the presence of sulfur on other particle surfaces. A short trend from the silicate corner in the diagram (Mg+Fe+Si+K–Na–Cl) (Fig. 12 c) toward the halite composition indicates mixed analyses between these phases.



**Figure 12:** Ternary diagrams : (a) (Mg+Fe+Si+K–Al–Ca), (b) (Mg+Fe+Si+K–S–Ca), (c) (Mg+Fe+Si+K–S–Ca).

## 4. Discussion



## 291 4.1 Dust composition

292 Dust from potential source areas (PSA) has been characterized by bulk chemistry (Kandler et al., 2007) and mineralogy  
 293 (Scheuven et al., 2013). Mineralogical fingerprints include phyllosilicates, such as the ratio of illite to kaolinite concentrations  
 294 (I/K) (Caquineau et al., 1998; Chester and Johnson, 1971; Scheuven et al., 2013). The general trends are increases in I/K from  
 295 S to N and from E to W. The value for the present samples is approximately 2.9, consistent with values reported from the  
 296 Western Sahara, e.g., >2 in NW Algeria and Morocco. High carbonate contents (>10 wt.%, calcite > dolomite) are also a  
 297 fingerprint of dust from the northwestern part of Africa (Avila et al., 1997). Similar to illite, carbonate content decreases  
 298 continuously toward the south (Paquet, 1984). The high carbonate content of dust emitted from the northern regions within  
 299 and adjacent to the Atlas Mountains reflects the geology of those regions (Grousset et al., 1992).

300

## 301 4.2 Iberulite formation and internal structure

302 The IF on February 6th in Fribourg was observed in person. The meteorological conditions during the fall are therefore well  
 303 known. Two mechanisms have been proposed for aggregating a large number of dust particles within a drop (Diaz-Hernandez  
 304 and Sanchez-Navas, 2016; Párraga et al., 2021): 1. The coalescence of drops within a cloud increases the number of  
 305 condensation nuclei within a single growing drop, i.e., in-cloud scavenging (ICS) (also denoted as 'nucleation scavenging'), or  
 306 2. the concentration increases through collisions between the particles and a drop below the cloud along its trajectory to the  
 307 ground. This mechanism is called below-cloud scavenging (BCS) (also denoted as impaction scavenging) (Pruppacher and  
 308 Klett, 2010). The BCS rate strongly depends on rainfall intensity, raindrop size distribution, and the drop's ability to collect  
 309 particles, called the collection efficiency (CE) (Volken and Schumann, 1993; Slinn, 1977; Laakso et al., 2003).  
 310 CE is the number (or mass) of particles captured by a drop of a given size relative to the particle number in the cylindrical  
 311 volume swept by the drop per unit trajectory length. If CE were independent of the properties of the drop, the particle,  
 312 environmental parameters, and the capture processes, the grain-size distribution in the iberulite would reflect the size  
 313 distribution of the dust in the volume traversed by the drop. However, this is not the case (see Fig. 10). The mechanisms  
 314 contributing to the capture process depend on particle size and the fluid flow around the falling droplet.  
 315 If the drop with a radius  $R$  falls vertically, e.g., no deviation by wind, the maximum number of possible collisions  $N$  along a  
 316 trajectory  $l$  within the dust layer with mass density  $M_d$  and particle density  $d$  can be calculated:

317

$$318 \quad N = \frac{M_d \cdot 2\pi R^2 \cdot l}{\frac{4}{3}\pi R^3 \cdot d} \quad (4)$$

319

320 For particles >1µm in diameter colliding head- on with a droplet, bouncing is unlikely, and the sticking efficiency is taken as  
 321 100% (Pruppacher and Klett, 2010). However, for particles with radii between 0.3 µm and 1µm, the CE, defined as the ratio  
 322 of the total number of collisions given by eq. 4 to the number of particles captured by the droplet, is < 1. This is due to the





323 airflow around the falling droplet. Particles within this size range, despite being on collision trajectories, follow the flow lines  
 324 and are carried around the droplet. Larger particles, due to their inertia, cross the flow lines and impact the droplet, resulting  
 325 in  $CE = 1$  (capture by impaction). Small particles ( $< 0.1 \mu\text{m}$ ) that follow the flow lines may be pushed by Brownian motion  
 326 into the droplet's wake and deposited on its rear end (Brownian capture), and  $CE$  is also close to 1. Capture may also be due  
 327 to thermophoretic and diffusiophoretic forces. Thermophoretic forces "push" aerosol particles toward a frozen drop during  
 328 evaporation and away from it during vapor deposition. During diffusive crystal growth, a flow known as Stefan's flow exists  
 329 near the hydrometeor surface, exerting a force on nearby aerosol particles that pulls them toward the surface (Santachiara et  
 330 al., 2023). Particles, independent of size, may follow the flow lines, but at some point the flow lines are closer to the drop's  
 331 surface than the particle's radius; e.g., the particle meets the drop's surface and sticks to it (capture by interception). This  
 332 process has a rather low capture efficiency. For particles with radii between  $0.3$  and  $1.0 \mu\text{m}$ , none of the above-described  
 333 capture processes is effective, and  $CE$  decreases by two orders of magnitude. This decrease in  $CE$  was first described by  
 334 Greenfield (Greenfield, 1957), and the lack of particles in the  $0.3 - 1.0 \mu\text{m}$  range is, therefore, referred to as the Greenfield  
 335 gap. Comparing the PSD of the free aerosol - we took the JSF PSD as representative for the distribution for the PSD inside the  
 336 Saharan dust cloud - and the one inside the iberulite, a shift of the maximum to larger sizes in the PSD inside the iberulite is  
 337 apparent, e.g., the presence of a Greenfield gap in the expected range (see Fig. 10).  
 338 Although no precipitation was observed on the ground during the afternoon of February 6th, 2021, some iberulites striking car  
 339 windshields were still wet, indicating that rainfall was present along the trajectory but had largely evaporated before reaching  
 340 the ground. The dust concentration distribution from the ceilogram taken on the day of the iberulite fall in Payerne shows three  
 341 maxima between  $4500$  m altitude and the ground. The dust concentration maxima within the cloud and just above the ground  
 342 are less significant than the massive layer between  $3000$  m and  $1000$  m AGL.  
 343 The temperature at the upper boundary of the dust layer was below  $0^\circ\text{C}$  and turned positive around  $2500$  m. However, melting  
 344 of the frozen hydrometeors most likely began at a much lower level due to the low relative humidity, in which the water vapor  
 345 density of the surrounding air is lower than the equilibrium water vapor density of frozen hydrometeors at the freezing point;  
 346 therefore, sublimation of water vapor occurs from the surface of the latter. Because of this sublimation process, latent cooling  
 347 can push the surface temperature of the hydrometeor below  $0^\circ\text{C}$  (Matsuo and Sasyo, 1981; Heymsfield et al., 2015). The  
 348 width of the non-melting layer in the atmosphere increases nearly linearly with decreasing relative humidity, from  $120$  m at  
 349  $90\%$  to  $700$  m at  $50\%$ . The relative humidity at the top level of the main Sahara dust layer is around  $70\%$  and decreases with  
 350 decreasing altitude. It is therefore probable that the melting level is displaced far into the massive Saharan dust layer.  
 351 Although difficult to distinguish from aerosol particles, the reflections just below the cloud base may originate from frozen  
 352 hydrometeors. Both solid ice crystals and liquid drops can capture particles. Frozen hydrometeors can occur as small particles,  
 353 columnar or planar crystals (branched = snowflakes), combinations of these, aggregates, and graupel, all with varying degrees  
 354 of riming (Magono and Lee, 1966; Grazioli et al., 2015; Praz et al., 2017). Scavenging of aerosol particles by frozen  
 355 hydrometeors is more complex than by raindrops because of their non-spherical shapes and falling behavior. With increasing  
 356 Reynolds number and shape irregularity, the falling motion of frozen particles transitions from steady to periodically changing



(fluttering, spiraling, zigzagging) and chaotic trajectories (Hashino et al., 2016; Kajikawa, 1992; Tagliavini et al., 2021; Mccorquodale and Westbrook, 2021; Stringham et al., 1969). Columnar ice particles align their long axis perpendicular to the (vertical) fall direction. Hexagonal ice plates and irregular snowflakes adopt a broadside orientation relative to the flow direction. Flow around such frozen hydrometeors is characterized by a deviating pattern around the front (Martin et al., 1980) and by eddies in the rear of the falling hydrometeor (Hashino et al., 2016; Wang et al., 2021). The distribution of dust particles (diameter 0.2 – 2.0  $\mu\text{m}$ ) on hexagonal ice plates shows increased concentration along the edges and corners (Prodi, 1976), whereas particles on snowflakes are distributed inhomogeneously (Ma et al., 2013).

Several studies on BSC (Feng, 2009; Kyrö et al., 2009; Paramonov et al., 2011) indicate that frozen hydrometeors are more effective scavengers of aerosol particles than rain on an equivalent water-content basis. The fluttering, zigzagging fall style of frozen hydrometeors prolongs their trajectories, thereby increasing the probability of collisions with aerosol particles and, consequently, CE. CE also increases with temperature and relative humidity. The highest CE has been recorded for mixed precipitation types and frozen hydrometeors at temperatures slightly above 0 °C. At these temperatures, frozen hydrometeors develop a stickier liquid layer at the surface. The increase in CE at higher RH is attributable to increased collector capacitance (Paramonov et al., 2011). The nature and size distribution of frozen hydrometeors also influence CE (Jung et al., 2015). CE decreases with increasing snowflake diameter, whereas dendrites and columns do not show this decreasing tendency. The largest CE was observed for columnar ice particles. The Greenfield gap in snowfall situations was in a smaller size range of 0.09-0.3  $\mu\text{m}$  compared to the gap observed for rain. This range is also observed for the iberulites collected in the present fieldwork, supporting frozen hydrometeors as dust scavengers. The increased CE of frozen hydrometeors is probably also the reason for the larger diameters observed in Swiss iberulites compared to Iberian iberulites.

When frozen hydrometeors exit the non-melting zone, for example, by crossing the melting level, they melt or evaporate. They exhibit a wide range of melting behaviors (Oraltay and Hallett, 2005). Ice crystals melt from the outside inward and are covered with a water film. Columnar ice particles melt uniformly, forming a thin liquid film that, as melting continues, bunches into one or several liquid bulges along the column (Knight, 1979; Fujiyoshi, 1986). Further melting within the bulges is slower, and the bulges coalesce into a single spherical drop containing the remaining ice particles. In melting snowflakes, liquid droplets gather preferentially in concave regions at the intersection of dendrites. When the concave areas are filled, the meltwater flows out, forming a spherical drop around the leftover ice particles (Oraltay and Hallett, 2005; Leinonen and Lerber, 2018). In experiments using a levitation test rig to study the melting process, a single ice crystal became spherical at 68.6% of melting, independent of its initial shape (Yan and Palacios, 2024).

Field observations (Heymsfield et al., 2015) spanned the smallest to the largest particles. In contrast to earlier studies (Ohtake, 1969; Barthazy et al., 1998), the largest particle size increased through the melting layer due to aggregation (Mcfarquhar et al., 2007), because the particles become stickier due to the liquid layer covering their surfaces. The particle concentration did not increase, suggesting that particle breakup during the melting process is not significant. The behavior of scavenged aerosol particles within melting hydrometeors has not been reported to date. Most likely, they are included in the newly formed raindrop.



391 Within and below the melting layer, due to the very low relative humidity (45% at 1000 m, at the bottom of the massive dust  
392 layer), raindrops will dry very rapidly. A technological analogue of this process is the (spray) drying of a powder suspension,  
393 which is used, e.g., in the ceramic, food, or pharmaceutical industry (Santos et al., 2018). The difference between the drying  
394 of a droplet in a spray drier and that of a rain droplet falling out of a cloud is the temperature difference between the droplet  
395 and the drying air stream, which is around 200°C in a spray drier. Droplets in a spray dryer relax into spherical shapes and are  
396 analogous to raindrops.

397 The latent heat of water evaporation induces a strong heat flux from the droplet surface into the gas stream. This, in turn, drives  
398 thermophoretic particle displacement toward the droplet surface and a specific microcirculation within a layer near the surface,  
399 driven by the surface tension gradient (Iskandar et al., 2003). This microcirculation entrains particles toward the droplet  
400 surface, leading to size- and density-based segregation. Smaller and/or less dense particles move faster toward the droplet  
401 surface, forming a thick surface layer. Coarser particles are concentrated in the core. Such a shell-core structure was observed  
402 in the present iberulite and has also been reported in the Iberian examples (Díaz-Hernández and Párraga, 2008; Párraga et al.,  
403 2021), with fine-grained phyllosilicates concentrated in the shell and coarse-grained quartz grains in the core.

404

## 405 5. Conclusion

406 To our knowledge, this analysis represents the first detailed study of iberulite fall in Switzerland. In contrast to the Iberian  
407 cases, the IF fall occurred under much colder conditions, e.g., temperatures around 0° and decreasing relative humidity along  
408 the fall trajectory. Iberulites form through the accumulation of particles inside raindrops. A comparison of the PSD inside the  
409 iberulites with the PSD measured in the dust cloud at the Jungfraujoch, which encountered the same cloud, revealed a  
410 Greenfield gap in the iberulite PSD, i.e., a lack of particles in the range between 0.1 and 0.3 µm. The presence of a Greenfield  
411 gap within the iberulites supports below-cloud scavenging, i.e., particle capture through collisions of particles with droplets  
412 below a cloud, as the mechanism for particle accumulation. The gap arises from particle trajectories in the flow around the  
413 falling droplet, which depend on particle size. The freezing temperature, low relative humidity, and ceilogram signals suggest  
414 that particle collisions occurred not with raindrops but with snowflakes or ice crystals. The latter are known to be better  
415 scavengers than raindrops.

416 Since 2021, more iberulites have been found in filter samples collected during SDEs in Switzerland. They will be analysed to  
417 confirm the findings and to gain a better understanding of the relationship between iberulite formation and meteorological  
418 conditions.



## 419 Author contributions

420 BG designed the study. CN, BG, and JR performed the measurements. BG analyzed the data. MS provided data from  
 421 Jungfraujoch. BG, JR, and DJ wrote the paper. All co-authors proofread and commented on the paper.

## 422 Competing interests

423 The authors declare that they have no conflict of interest..

## 424 Acknowledgements

425 The authors acknowledge Maxime Hervo from Meteoswiss for the supply of meteorological and lidar data. We thank also  
 426 Mario Wannier for supplying iberulites collected in St. Léger. NOAA for HYSPLIT? SEVIRI? Originator of Figure 2? PSI  
 427 for SMPS data from JFJ.

## 428 Financial support

429 This research received no external funding.

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