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Exploring size-dependent dynamics of photosynthetic cells in rainwater: the influence of atmospheric variables and rain characteristics

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Abstract

20 The presence of microalgae in the atmosphere raises health and environmental concerns. Despite recent scientific advances, our knowledge of the origins and dynamics of photosynthetic cells in relation to atmospheric processes is limited due to a lack of empirical data. To address this gap, we conducted a one-year survey, collecting and analyzing rainwater samples. This study proposes to investigate the temporal dynamics of photosynthetic cells based on their size in combination with a unique dataset of variables of interest: type of rain and its characteristics, local meteorology, 25 concentrations of inorganic chemical species, and long-range air mass transport. The analysis of the biochemical composition of rainwater, along with its correlation with the origin of air masses using ions as tracers, provides evidence of the long-range transport of photosynthetic cells. Additionally, our study reveals distinct removal mechanisms from the atmosphere for photosynthetic cells depending on their size. Our results suggest that convective events with high-intensity rainfall led to an efficient 30 removal of medium-sized photosynthetic cells (4-15 μm) from the atmosphere. However, removal mechanisms for small (<4 μm) and large-sized cells (>15 μm) are not influenced by microphysical rainfall characteristics and seem govern by different atmospheric processes: dry deposition is proposed to be a significant mechanism for the removal of large-sized photosynthetic cells, while small-sized cells detected in rain is correlated with the horizontal wind speed and duration of rainfall, 35 particularly during stratiform events. This implies that the removal of photosynthetic cells from the atmosphere is strongly influenced by environmental variables, which are expected to vary in response to global change. Therefore, it is crucial to enhance the monitoring of photosynthetic cells in relation

to atmospheric processes and investigate the potential impact of the dissemination of genetic material from distant sources on recipient ecosystems.

40 1. Introduction

The recent discovery of bioaerosols in various atmospheric compartments, such as air, cloud, and rainwater (Wiśniewska et al., 2019; Després et al., 2012a; Delort et al., 2010; Cheng et al., 2021) raises concerns about their potential impact on atmospheric processes (Deguillaume et al., 2008; Morris et al., 2011; Möhler et al., 2007), as well their effect on health and potential economic impacts (Jalasvuori, 2020; Wiśniewska et al., 2019). Photosynthetic cells (PS), including cyanobacteria, chlorophytes, and diatoms, have been sporadically monitored in the atmosphere, in comparison to bacteria, fungi, and viruses (Sharma et al., 2007; Genitsaris et al., 2011). The widespread presence of cyanobacteria and eukaryotic microalgae in the atmosphere, albeit in lower abundance than bacteria, fungi, and viruses, highlights their potential dissemination over long distances (Gaylarde and Gaylarde, 2000; Rosas et al., 1989; Sharma et al., 2006; Curren and Leong, 2020; Murby and Haney, 2016; Genitsaris et al., 2011). However, there is limited understanding of the factors that contribute to the widespread distribution of these microorganisms in the atmosphere (Burrows et al., 2009; Fröhlich-Nowoisky et al., 2016; Šantl-Temkiv et al., 2022; Tesson and Šantl-Temkiv, 2018). Different environmental variables, such as season, atmospheric temperature, wind speed, and relative humidity, have been suggested to explain the dynamics of microalgae without showing a clear pattern, possibly due to the influence of multiple emission sources (Sharma et al., 2007; Genitsaris et al., 2011). Marine areas represent a significant sources of airborne microalgae (Lewandowska et al., 2017) and sea spray aerosols emitted from marine surface can transport airborne microalgae up to 1000 km inland (May et al., 2018; Manders et al., 2010). Splashing by precipitation on leaves and buildings can also lead to the emission of airborne microalgae (Gaylarde and Gaylarde, 2000; Fröhlich-Nowoisky et al., 2016), introducing a continental potential origin for these PS cells with both long range origin and local one. The wet atmospheric deposition mechanisms have been well-documented through field and laboratory experiments using black carbon, radioactive particles from nuclear accidents and pollutants, as well as numerical and theoretical approaches (Wang et al., 2010; Andronache, 2003; Mircea et al., 2000; Quérel et al., 2014). However, the removal of bioaerosols from the atmosphere through the wet deposition mechanisms is currently not well understood, although understanding these processes is crucial as they influence the lifespan of microalgae in the air and their ability to travel over long distances. Two types of wet deposition have been highlighted: in-cloud and below-cloud scavenging (Slinn, 1976; Rasch et al., 2000). In-cloud scavenging is related to activation/nucleation processes since aerosols can serve as cloud condensation nuclei (CCN) and ice nucleation (IN); photosynthetic cells are known to participate to cloud droplet and crystal formation (Després et al., 2012b; Möhler et al., 2007; Morris et al., 2014; Christner et al., 2008; Tesson and Šantl-Temkiv, 2018; Bowers et al., 2009). This process may increase the chances of microalgae survival by shortening their lifespan in the atmosphere. The other wet deposition process, refer as the below-cloud scavenging, occurring when aerosols are removed from the atmosphere due to collection by falling raindrops. The scavenging coefficient in this process is influenced by microphysical variables (rain rate and raindrop

size distribution), the aerosol size (Andronache, 2003; Mircea et al., 2000; Quérel et al., 2014; André et al., 2007; Hou et al., 2018; Möhler et al., 2007; Blanco-Alegre et al., 2022), as well as the type of precipitation (convective and stratiform) (Huang et al., 2019; Wu et al., 2013; Niu et al., 2010; Llasat, 2001). For example, convective events, characterized by higher turbulence and rapid exchanges between the cloud and underlying air (Jaffrezo et al., 1990; Pruppacher and Klett, 2010) may enhance the particle collection efficiency and thus improve the removal of aerosols from the atmosphere.

Eukaryotic microalgae and cyanobacteria, which have sizes ranging from 0.2 to several micrometers, have traditionally been believed to have low removal rates according to microphysical modeling. This phenomenon has been referred to as the "scavenging gap" (Andronache, 2003; Greenfield, 1957; Ohata et al., 2016). However, recent studies have reported higher scavenging efficiencies for small-sized microbial aerosols compared to the predicted values from microphysical modeling (Moore et al., 2020; Chate, 2005). The increased scavenging efficiency for microbial aerosols in the size range of 0.2 to 2 μm has been attributed to raindrop diameter (Moore et al., 2020). Furthermore, an increase in concentration of photosynthetic cells (PS) in rainwater have been reported, particularly during rain events characterized by larger raindrop diameters, lower air mass altitudes, and a lower position of the bright band in the cloud (Noirmain et al., 2022).

The size-dependent dynamics of photosynthetic cells and their removal from the atmosphere through atmospheric processes, particularly in relation to different rain intensities, have not been thoroughly investigated (Luan et al., 2019; Moore et al., 2020). However, with the anticipated rise in temperatures and rainfall intensity due to global changes, the dynamics of bioaerosols are expected to undergo changes. It is projected that global warming will lead to increased phytoplankton biomass and blooms worldwide (Plaas and Paerl, 2021; Jöhnk et al., 2008; Richardson et al., 2019; Bartosiewicz et al., 2019), resulting in an increased release of photosynthetic (PS) cells into the atmosphere (Curren and Leong, 2020; Genitsaris et al., 2011) and subsequent dissemination into ecosystems. The dispersion of these bioaerosols raises ecological concerns that must be carefully considered (Kokko and López-Sepulcre, 2006).

The present work aims to study the dynamics of PS cells in rainwater and to analyze how the type of rain events, the meteorological variables, and the air masses origins influence the size-dependent dynamic of photosynthetic cells, including the wet atmospheric deposition. To achieve this goal, we developed an original interdisciplinary approach operated at the Aydat instrumental site in France. The physical properties of rain events, the meteorological conditions, and the air masses origins have been characterized in parallel with the dynamic of photosynthetic cells in rainwaters. 28 rain events composed of one or several intra-samples were collected from September 2020 until October 2021 using a rain sampler to measure the concentrations of photosynthetic cells and ions. The sequential analysis of photosynthetic cells during rain events provides a better understanding of their dynamics based on their size and pigment composition. Cloud and rain parameters are measured using an advanced radar instrumental suite, allowing to identify the type of precipitation (convective vs stratiform), in association with local meteorology and long-range air mass origins.

115 2. Materials and methods

2.1. Atmospheric instrumental setup

The experimental setup has been described previously in Noirmain et al. (2022). It includes a cloud radar (Metek Mira35c) and a disdrometer (Parsivel²). The cloud radar provides information on various cloud parameters such as cloud altitude, height of the bright band, cloud reflectivity, and reflectivity streaks. In a complementary way, the disdrometer is utilized to measure the size distribution of raindrops, falling speed, peak rainfall intensity, mean rain rate, and average drop diameter.

The instruments have been installed in 2020 at Aydat, a commune located in France (45.66°N; 2.98°E), at the altitude of 850 m above sea level. The air temperature, wind speed, and relative humidity are also continuously monitored by a weather station operated by the French meteorological network "Météo France" and located at Saint-Genès-Champanelle (45.71°N; 3.01°E), approximately 7 km northeast of Aydat, at 893 m above sea level.

2.2. Sampling of rainwater

Rain samples have been collected over a one-year period from September 2020 to October 2021 using an automatic sequential sampler (EIGENBRODT GmbH & Co. KG, Baurat-Wiese-Straße, Königsmoor) designed for high temporal resolution rain collection. Two rain events are considered separate if they are interrupted by more than 15 minutes without rain. In such cases, the second rain event is collected in a new bottle. Each rain bottle has a capacity of 250 mL, and if the rain event exceeds this volume, intra-samples are collected by replacing the full bottle with a new one. This sequential sampling approach was previously employed to describe scavenging processes due to its high temporal-resolution (Jaffrezo et al., 1990; Noirmain et al., 2022). Snow samples are excluded from the analysis since aerosol scavenging by snow involves different mechanism than that induced by the rain; this could induce biases in the analysis of the wet deposition mechanisms.

The polyethylene funnel is thoroughly cleaned before each rain event using detergent and sterilized Milli-Q water to eliminate any potential contamination between rain events. The rainwater samples are stored at 4°C in the dark and analyzed within 48 hours after the end of the rain event.

2.3. Chemical and biological analysis of rainwater

The number of photosynthetic cells is quantified using the flow cytometry protocol detailed in Noirmain et al. (2022). Fresh rain samples are pre-concentrated through ultra-fast filtration with a Vivaspin® 100 kDa filter. Autofluorescence signals from photosynthetic pigments are detected using bandpass filters and a 640 nm and 488 nm laser, with emissions collected through FL3, FL2, and FL4 detectors (using 670 Long Pass, 585/42, and 661/16 Band Pass filters, respectively) on a BD FACSCalibur™ machine. A flow cytometry size calibration kit (F-13839) is utilized, which consisted of six suspensions of unstained polystyrene microspheres with known diameters of 1, 2, 4, 6, 10, and 15 µm. The microspheres are passed through the machine following the manufacturer's instructions to set specific size ranges (Givan, 2013). Intervals of [1-2], [2-4], [4-6], [6-10], and [10-15] µm are defined, along with supplementary ranges below 1 µm ([<1]) and above 15 µm ([>15]). The population labeled as "phycoerythrin" enables to detect pink cyanobacteria and Cryptophyceae (Read

et al., 2014). The "chlorophyll" population is capable of detecting Chlorophyceae, diatoms, and Chrysophyceae, while the "phycocyanin" population allows us to detect blue-green cyanobacteria (Noirmain et al., 2022).

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The acquisition is performed in high flow rate mode for 60 s with adjusted thresholds on FSC and FL3.

We calculate the wet concentration of photosynthetic cells using the following formulae:

$$C_w = \left(\frac{E \times 10^6}{F \times t} \right) / a$$

Where C_w refers to the wet concentrations of photosynthetic cells (cells L⁻¹), calculated from E the number of events (i.e., the number of photosynthetic cells that pass through the laser) recorded by flow cytometry, F the exact flow rate determined using Milli-Q water for each rain sample through the acquisition time and the gravimetric loss from the water sample (μl min⁻¹) and t the acquisition time (min). The resulting concentration is then adjusted by a the concentration factor determined by volumetric loss from the rain sample after ultra-fast filtration.

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The remaining samples are filtered through a 0.2 μm Nylon membrane pre-rinsed with 500 ml of Milli-Q water to eliminate microorganisms, the pH is measured on fresh samples and the remaining is stored at -20°C until analysis by ion chromatography. The wet concentrations (C_w) of dissolved nutrients, including Ca²⁺, Cl⁻, K⁺, Mg²⁺, Na⁺, NH₄⁺, NO₃⁻, PO₄³⁻, and SO₄²⁻ in the rainwater are analyzed using ion chromatography (IC) on a Dionex ICS6000 system. An IonPac AG11-HC (guard-column 2 × 50 mm) and an IonPac AS11-HC 260 (analytical column 2 × 250 mm) are used for the analysis of anions, while an IonPac CG-16 (guard-column 2 × 50 mm) and an IonPac CS16 (analytical column 2 × 250 mm) are utilized for the analysis of cations. The elution is performed in gradient mode using KOH (1 mM to 60 mM in 35 minutes, flow rate of 0.36 ml min⁻¹) for anions and isocratic mode with MSA (methanesulfonic acid at 30 mM, flow rate of 0.25 ml min⁻¹) for cations. The chromatograms are recorded using a conductimetric cell detector and analyzed with Chromeleon 7.2 software. Three replicate measurements are conducted to determine the nutrient concentration. This allows calculating standard deviations illustrated on Figure 1 in SI. The limit of detection (LOD) is estimated with a signal-to-noise ratio of 3:1, while the limit of quantification (LOQ) is obtained using signal-to-noise ratio of 9:1 (Shrivastava and Gupta, 2011). Calibration curves are also generated for each ion to validate the limits of detection (LOD) and limits of quantification (LOQ) obtained through the signal-to-noise method. For anions, the LOD ranged from 0.7 μg L⁻¹ for PO₄³⁻ to 2.3 μg L⁻¹ for Cl⁻ and the corresponding LOQ ranged from 2.1 to 6.8 μg L⁻¹, depending on the specific anions (Table S1).

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For cations, the LOD ranges from 0.7 for K⁺ to 141 μg L⁻¹ for Mg²⁺, the corresponding LOQ ranged from 2.1 μg L⁻¹ to 425 μg L⁻¹, depending on the specific cations (Table S1).

2.4. Evaluation of the wet atmospheric deposition fluxes

The wet deposition fluxes (F_w) of different ions and photosynthetic cells are calculated using the following formulae (Milinković et al., 2022):

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$$F_w = (C_w \times V)/(s \times t)$$

Where F_w refers to the wet deposition flux for the ions and photosynthetic cells ($\text{mg m}^{-2} \text{h}^{-1}$ and $\text{cells m}^{-2} \text{h}^{-1}$, respectively), calculated from C_w the concentration of ions and photosynthetic cells in the rainwater (mg L^{-1} and cells L^{-1}) during each intra-events according to V the volume of rainfall, t the duration of the sampling period (hours) and s , the surface area of the funnel collecting the rain which is equal to 0.05 m^2 .

2.5. Physical and meteorological data

2.5.1. Air mass trajectories

Back trajectories are calculated for each precipitation event starting from the altitudes of the cloud layers observed on the Mira35 cloud radar vertical profiles. The back-trajectories have been calculated with CAT (Computing Atmospheric Trajectory tool; Baray et al., 2020), a 3D forward/backward kinematic trajectory model initialized with wind fields from the ECMWF ERA-5 climate reanalysis. The wind fields are extracted every 3 h with a spatial resolution of 0.25° in latitude and longitude, and at 29 pressure levels between 50 and 1000 hPa. CAT uses bilinear interpolation for horizontal wind fields and time and log-linear interpolation for vertical wind fields with a temporal resolution of 15 min and a total duration of 120 h.

The geographic origin is determined by counting the number of trajectory points in 9 pre-defined sectors (NNE, NEE, SEE, SSE, SSW, SWW, NWW, NNW, and NEAR) and points over land and sea, separated into those close to the ground (below 2 km) and those in the free troposphere (above 2 km). The CAT model has been initially developed to establish the atmospheric composition measurement footprint at the puy de Dôme high-altitude observatory (Baray et al., 2020). It has been applied in conjunction with chemical measurements to classify clouds at this atmospheric station (Renard et al., 2020).

2.5.2. Classification of precipitation events

The classification of precipitation events is based on the meteorological nature of the cloud mass and the intensity and variability of macro- and microphysical rain characteristics, including drop diameter, rain rate peak, and cloud reflectivity. To accurately classify the type of precipitation, we employ multiple criteria, rather than relying on a single physical variable which can underestimate the number of events classified to each rain type (Huang et al., 2019; Wu et al., 2013; Niu et al., 2010; Llasat, 2001). Furthermore, these criteria are analyzed throughout the duration of the events. In stratiform events, there is a tendency for the mean drop diameter to decrease when the rain rate intensity exceeds 10 mm h^{-1} . Conversely, in convective events, the mean drop diameter tends to increase with a higher degree of variability (expressed as standard deviation - SD) as the rain rate increases during the course of the event (Niu et al., 2010). We used the following thresholds to classify the events (Niu et al., 2010; Steiner et al., 1995):

- The thresholds for the peak of rainfall intensity are set at $10 \pm 1.5 \text{ mm h}^{-1}$ standard deviations (SD);

- The thresholds for the mean drop diameter, normalized by the total number of raindrops, is set at 0.5 mm;
- The mean cloud radar reflectivity (Z factor) threshold is set at 30 dBZ;
- The reflectivity during each intra-event, recorded from the cloud radar, is analyzed to assess the presence of reflectivity streaks with rain bands, which are associated with convective events.

An event is categorized as convective when all thresholds are exceeded.

2.6. Statistical analysis

A Redundancy Analysis (RDA) is performed to evaluate the statistical relationships between the meteorological variables and rain microphysical characteristics (explanatory variables) and the fluxes or concentrations of photosynthetic cells and inorganic ions (response variables) (Legendre and Legendre, 2012; Rellstab et al., 2015; Forester et al., 2017). The significance of this model is evaluated through permutations tests, which help identifying significant predictors in the data matrix using the Akaike Information Criterion (AIC) in a stepwise algorithm (Oksanen, 2010).

The Spearman's rank correlation test is calculated to evaluate the significance of the correlations between the wet deposition fluxes (F_w) of PS cells and the rain rate; and between the wet concentrations (C_w) of PS cells and the wind speed, during stratiform and convective events.

3. Results

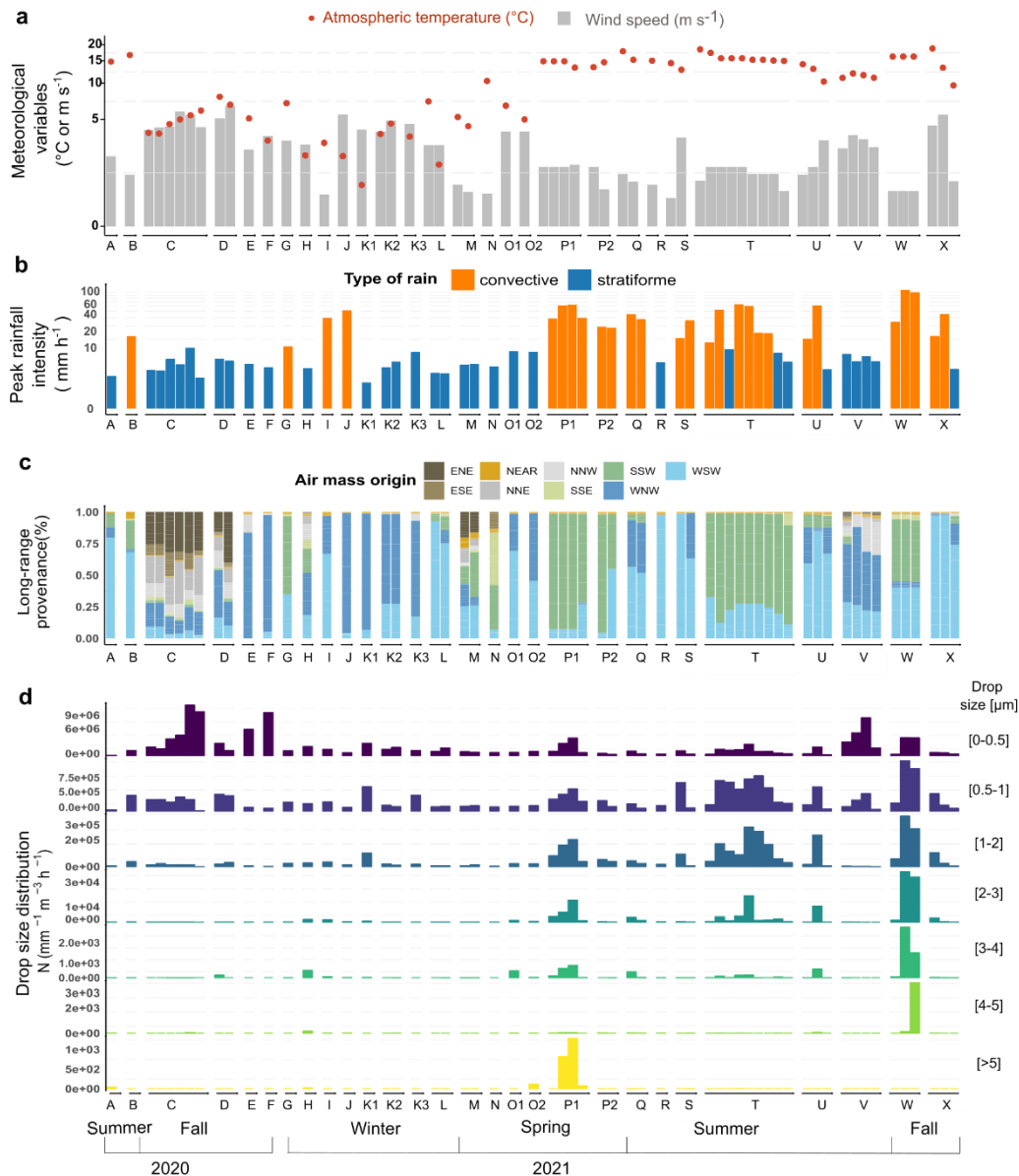
3.1. Dynamical context: air mass origins and characteristics of precipitation

Our study analyses 28 individual rain events, including 60 sequential intra-events distributed into 27 convective and 33 stratiform intra-events based on the criteria given in section 2.4.2. (Table S3). As expected, convective events are associated with significant higher temperatures and lower horizontal wind speeds than stratiform events (p -value = $1.17 \cdot 10^{-6}$ & $7 \cdot 10^{-4}$, respectively) (Fig. 1a & Table S3). Three events (T, U and X), contain both convective and stratiform intra-events and are called "mixed" events (Fig.1c and Table. S1). T presents the highest number of intra-events, composed of 6 convective and 3 stratiform events. Among the 60 sequential intra-events, 51% are composed of a single sample with a rain volume less than 250 ml and are primarily characterized as stratiform type. Stratiform events occur throughout different seasons, including fall, winter, and spring, whereas convective events are mainly observed in summer and fall. Interestingly, the types of events are often associated with specific air mass origins (Fig. 1b). The dominant air mass origin during convective events is the SSW sector (Mediterranean Basin) (p -value = $1.6 \cdot 10^{-19}$), while stratiform events are often linked to continental origins from the ENE, ESE, and WNW sectors, especially during events C, D, and V (Fig. 1b). Furthermore, no trajectory points are recorded in the NNE and NNW sectors during convective events, while a low occurrence is observed during stratiform events (Fig. 1 and Table S3). Conversely, both convective and stratiform events are associated with oceanic origin. The origin of the air mass appears to remain relatively consistent during intra-events within a rain event. In contrast,

265 the size distribution of raindrops can vary throughout the duration of both convective and stratiform events (Fig. 1d). Specifically, convective event P1 exhibits a noticeable variation in the size distribution of raindrops, with a low number of raindrops larger than 5 mm appearing during the events.

270 The analysis of the raindrop size distribution shows that during convective events, small-sized raindrops (0-0.5 mm) make up 76% of the distribution, while raindrops in the higher size range (0.5-1 mm) account for 18%. In contrast, stratiform events are predominantly composed of small-sized raindrops, accounting for 96% of the distribution.

275 Other atypical events are observed, such as event W which has the highest rain rate and a peak at 104 mm h⁻¹ and is composed of raindrops of different size ranges (Fig. 1). Among stratiform events, event C stands out with the longest duration of approximately 22 h (composed of 6 intra-events), with the highest number of raindrops ranging from 0 to 0.5 mm in diameter at the end of the event (intra-events 5 & 6) and presenting consistent air mass origins. On the other hand, event D is characterized by the highest wind speed during the study period, exceeding a mean of 7.5 m s⁻¹ during a one-hour interval.



280 **Fig. 1:** (a) Mean atmospheric temperature and wind speed at Saint-Genès-Champanelle, expressed in
 100 log-scale (b) backward trajectory percentage by geographic origin, calculated using the CAT model,
 110 (c) rainfall intensities, classified into convective or stratiform, expressed in log-scale and (d) temporal
 120 evolution of the number of raindrops (N) ($\text{mm}^{-1} \text{m}^{-3} \text{h}^{-1}$) according to the raindrop size ranges (μm),
 130 recorded from the Parsivel² sensor measured at Aydat instrumental site. Each bar in the graph
 140 represents a single rain event or an intra-sequence event which are labelled with a distinct alphabetic
 150 letter. Multiple rain events occurring on a same day, with at least a 20-min interval without rain
 160 between them, are distinguished by an extra number after the letter (Table S2).

3.2. Photosynthetic cells in rainwater: temporal dynamics of their fluxes and concentrations

285 The wet deposition fluxes (F_W) and wet concentrations (C_W) of photosynthetic cells (PS) present a
 290 high variability during the whole study period (Fig. 2). The most abundant PS cells are those rich in
 chlorophyll and phycoerythrin, with chlorophyll-rich cells contributing to approximately 56 and 33%
 of the total PS cells F_W and C_W , respectively and phycoerythrin-rich cells accounting for 43 and 67%

of the total PS cells F_W and C_W , respectively. Conversely, PS cells containing phycocyanin contribute to less than 1 and 0.3% of the total PS cells F_W and C_W for all pigments, respectively. The size distribution of cells also varies among pigment classes. Phycoerythrin-rich cells are predominantly large-sized, with sizes above 15 μm , accounting for 52% and 82% of the total F_W and C_W for this pigment, respectively. In contrast, both chlorophyll-rich cells and phycocyanin-rich cells exhibit a more even distribution across different size ranges (Fig. 2). Approximately 54% of the total F_W of phycocyanin and phycoerythrin-rich cells are characterized by medium-sized cells, ranging from 4-10 μm and 4-15 μm in size, respectively. On the other hand, the C_W of cells for both pigment classes are characterized by a higher proportion of small-sized cells, accounting for 26% and 27% of the cells within the 0-1 μm size range, for chlorophyll and phycocyanin-rich cells, respectively (Fig. 2).

We investigate the relationships between the dynamic of PS cells (F_W and C_W) and environmental variables through a redundancy analysis (RDA) (Fig.3). We report that microphysical characteristics of rain, meteorological variables, inorganic composition of rainwater, and air mass trajectories are significant variables in the RDA models conducted on PS cells rich in chlorophyll and phycoerythrin. The first constrained axis (RDA1) explains 36.3% and 37.5% of the temporal variability in F_W of PS cells rich in chlorophyll and phycoerythrin, (Fig. 3a-b) and 49.4% and 22.8% of the variability in F_W and C_W of chlorophyll-rich cells, respectively (Fig. 3c-d). Due to the low proportion of variance explaining the temporal variability of phycocyanin-rich cells obtained from the RDA (Suppl. Fig. 3) and the lack of significance on the first constrained axis (RDA1) when considering the F_W , we refrain from interpreting the results of the RDA to avoid potential misinterpretation. This limited explanatory power can be attributed to the low abundance of phycocyanin-rich cells in the rain sample.

The results from the RDA analysis provide evidence of strong correlations between the F_W of PS cells which include both chlorophyll and phycoerythrin pigments, and rainfall intensity as well as the size of raindrops during convective events (Fig.3a & c). These correlations are also observed for the F_W of phycocyanin-rich cells but are not significant (Suppl. Fig.2a). Additionally, Spearman correlations tests reveal that the strength of correlations between the rain rate and the F_W of PS cells, considering all pigments, vary according to the type of rain events (Suppl. Fig. 3a). Indeed, during convective events, stronger and significant correlations are found for medium-sized cells ranging from 4 to 15 μm , with the highest correlation observed for cells within the 6-10 μm size range ($r = 0.58$, $p\text{-value} = 7.9 \cdot 10^{-8}$), while weaker correlations are observed for smaller cell sizes ($<4 \mu\text{m}$) and larger cell sizes ($>15 \mu\text{m}$) (Suppl. Fig.3a). On the other hand, during stratiform events, the correlations between the F_W and rainfall intensity are generally weak for all size ranges ($r < 0.3$) (Suppl. Fig. 3b).

Surprisingly, the RDA reveals one specific relationship concerning the phycoerythrin-rich cells above 15 μm , which are inversely correlated with the rain rate and atmospheric temperature during convective and stratiform events G, I and K2 (Fig. 3c). These specific events are characterized by air masses originating from the African continent and the Atlantic Ocean, which have travelled at high altitudes (above 2 km).

The analysis of the variability of the C_W of PS cells through the RDA show correlations between the C_W of small-sized PS cells and the wind speed during stratiform events (Fig. 3b & d). A similar result is found with the C_W of phycocyanin-rich cells (Suppl. Fig.3b). Additionally, The Spearman's rank correlation test confirms the significance of the correlations between the C_W and the wind speed when considering all pigments, especially during stratiform events (Suppl. Fig.3c-d). The strength of correlation is more robust for size ranges from 1-2 μm during stratiform events ($R = 0.48$, $p\text{-value} = 3.8 \cdot 10^{-6}$) (Suppl. Fig 3d); while it is generally weak during convective events for all ranges ($R \leq 0.3$) (Suppl Fig. 3c). Additionally, there is no observed correlation between wind speed and PS cells larger than 15 μm during both convective and stratiform events ($R \leq 0.1$).

We also investigated the potential link between PS cells and air masses origins. Notably, a correlation is found between the C_W of chlorophyll-rich cells, with size above 4 μm , and the C_W of terrestrial ions (Ca^{2+} , Mg^{2+} , and NH_4^+) and marine ions (Na^+ and Cl^-), during convective and stratiform events associated with continental (SSW) and oceanic (WNW) origins respectively (Fig. 3b). Furthermore, the C_W of the small-sized cells rich in chlorophyll are found to be high during stratiform events with air masses from Northeast origins (ENE) (Fig. 3b). We also observed a correlation between the air mass transported over the African continent (SSW) and the C_W of PS cells containing phycoerythrin (2-15 μm & > 15 μm), as well as the C_W of terrestrial ions (Ca^{2+} , NO_3^- , Mg^{2+} , and NH_4^+) (Fig.3d). On the other hand, the C_W of the small-sized cells (0-1 μm) rich in phycoerythrin are correlated with oceanic origins and the C_W of Cl^- (Fig. 3d). Similar results are reported with the C_W of medium and large-sized cells containing phycocyanin (4-15 μm), showing correlations with continental origins (SSW) and, in the case of specific events, a local origin near Aydat (NEAR) (Suppl. Fig. 2b).

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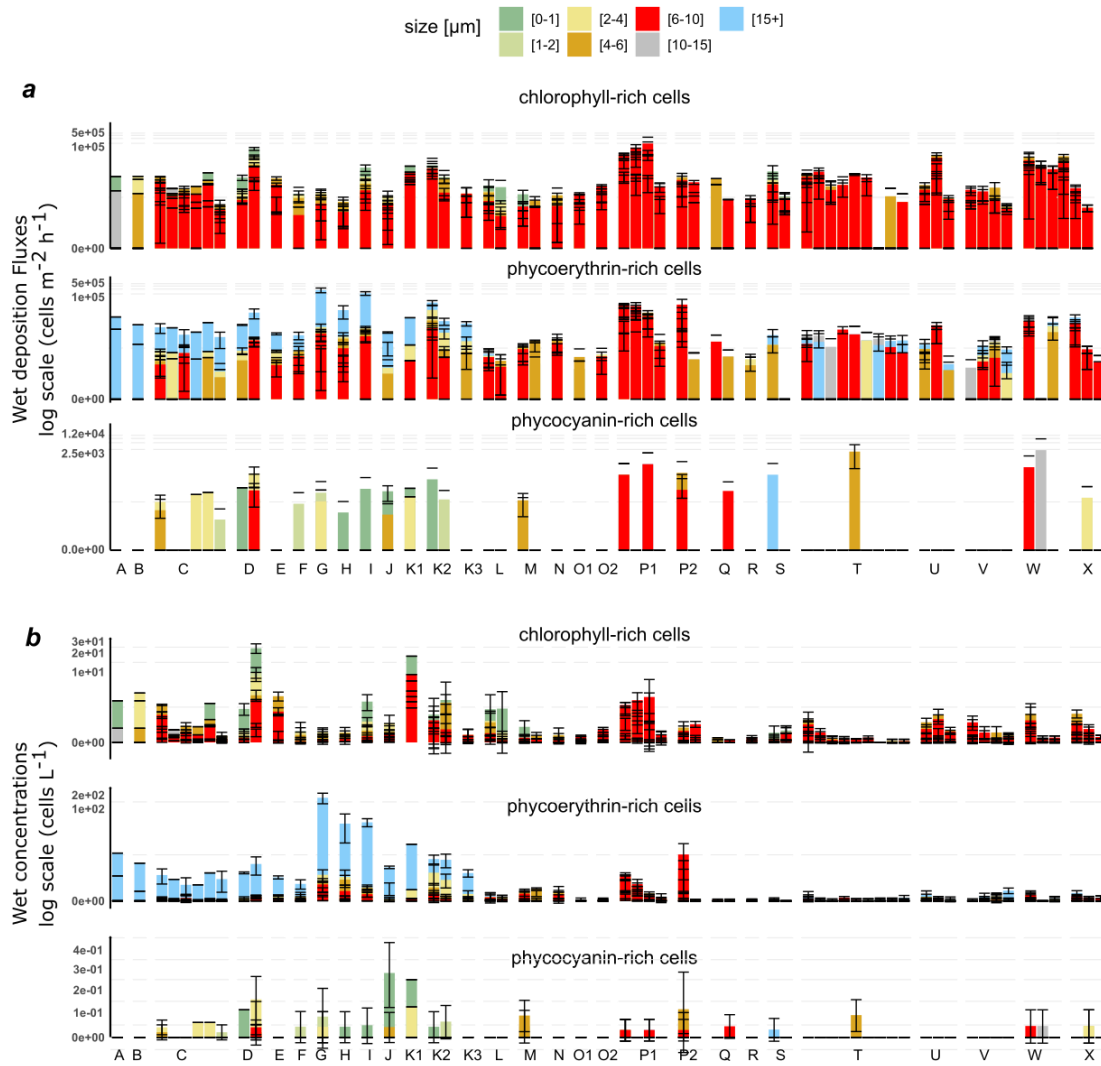


Fig. 2: Temporal variations of photosynthetic cells (a) wet deposition fluxes (F_W) ($\text{cells m}^{-2} \text{h}^{-1}$) and (b) wet concentrations (C_W) (cells L^{-1}) during the rain campaign, expressed in log-scale. Each bar in the graph represents a rain event or an intra-sequence event labelled with an alphabetic letter. Multiple rain events occurring on a same day, with at least a 15-min interval without rain between them, are distinguished by an added number after the letter.

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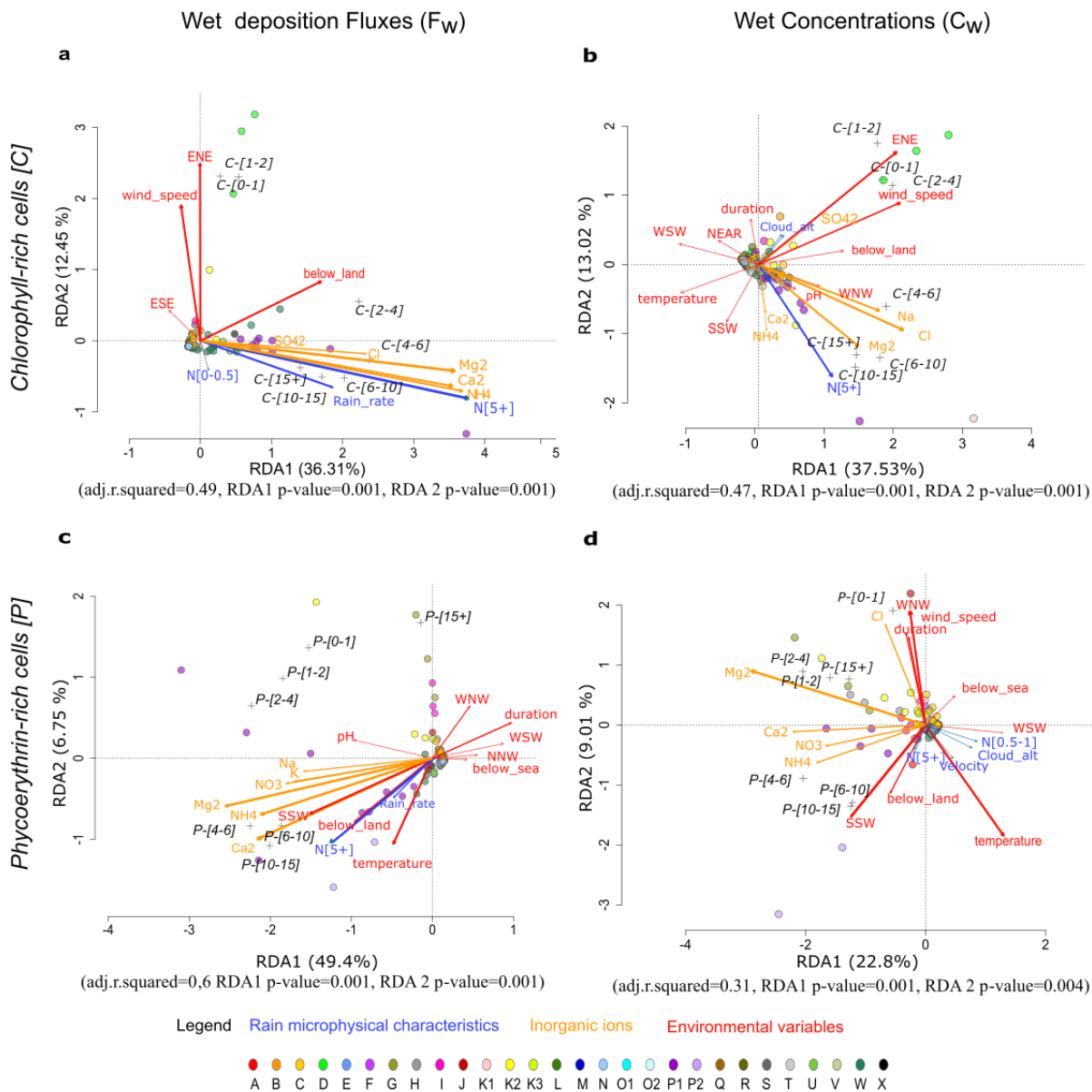


Fig 3: Redundancy Analysis (RDA) is performed using the wet deposition fluxes (F_W) and wet concentrations (C_W) of the PS cells rich in chlorophyll (C) (a & b, respectively) and phycoerythrin (P) (c & d, respectively), based on their cells size ranges ([0-1], [1-2], [2-4], [4-6], [6-10], [10-15], and [15+]) expressed in μm . The explanatory variables are represented in black italic font, while the significant predictor variables (selected by permutation tests) are displayed based on their classes: Blue represents rain microphysical characteristics, including terminal drop velocity ("velocity"), rain rate, cloud altitude ("cloud_alt"), and raindrop number (N) according to their range size ("N [0.5-1]" and "N [5+]" expressed in mm). Orange color represents ion concentrations, expressed in the same unit as the PS cells (expressed in $\text{cells m}^{-2} \text{h}^{-1}$ for the F_W and cells L^{-1} for the C_W). Red color represents environmental variables such as air mass origin traveling below 2 km over land and sea (referred to as "below-land" and "below-sea"), wind speed, air masses origins according to sectors ("ENE", "NNE", "ESE", "SSE", and "SSW"), and air temperature ("temperature"). The sequential rain intra-events are represented by circles, colored according to the rain events, and labeled with alphabetic letters.

4. Discussion

Predicting atmospheric dynamics of PS cells remains challenging due to the many processes involved related to origins, long-range transport, and wet deposition, controlling their selection and their viability. The analysis of our bio-physical-chemical dataset, characterizing 60 sequential rain events from highly contrasted atmospheric conditions (air mass origin, types of rain, and local weather), allows us to associate the dynamics of PS cells with specific rain event characteristics by discriminating for the first time the size of the PS cells and investigating their relationships with key environmental factors during both convective and stratiform events.

4.1. The dynamics of PS cells in rain is governed by different atmospheric processes according to their size

Our results suggest that the medium-sized (4-15 μm) PS cells are efficiently removed from the atmosphere by the rain, as evidenced by the strong correlations between the F_W of medium-sized PS cells and both the large-sized raindrop and rain rate (Fig. 3 b, d & f & Suppl. Fig. 3a). These variables significantly contribute to explaining a large portion of the variability in the F_W of medium-sized PS cells rich in chlorophyll and phycoerythrin-rich cells, respectively (Fig. 3a & c). Since these rain microphysical variables are known to play a role in the below-cloud scavenging process of bioaerosols (Andronache, 2003; Mircea et al., 2000; Hou et al., 2018; Möhler et al., 2007; Blanco-Alegre et al., 2022), it can be suggested that this process is significantly involved in removing the medium-sized PS cells from the atmosphere. This point is strengthened by the fact that stronger effect of this process is observed during convective events, compared to stratiform ones (Suppl. Fig.3a-b), probably due to the larger raindrop diameters and higher rain rates observed during convective events. In contrast to the medium-sized PS cells, the dynamics of the smaller ones ($<4\mu\text{m}$) exhibit a weak correlation with the rain rate during both convective and stratiform events (Fig. 3 & Suppl. Fig. 2). Furthermore, in our study, these small-sized PS cells account for only a small proportion of the total PS cell F_W (30 % of the total F_W), which could suggest that these small-sized PS cells experience a low below-cloud scavenging. This is actually reported for the small-sized aerosols particles (0.05-2 μm) referred as the "Scavenging gap" phenomenon (Greenfield, 1957; Andronache, 2003). This phenomenon suggests that these particles are too small for inertial impaction and too large for Brownian diffusion in order to significantly contribute to their deposition (Moore et al., 2020; Ohata et al., 2016). Quantifying the abundance of small-sized cells in the air and cloud water, and comparing them to their abundance in rainwater, would provide valuable insights into determining whether these cells can persist in the atmosphere for extended periods. Additionally, we report that the presence of these small-sized PS cells in rainwater is observed during events associated with high horizontal wind speed, particularly during stratiform events characterized by light rainfall intensities, low raindrop velocity, and extended durations. Since we report also a significant and strong correlation between wind speed and the C_W of small-sized PS cells during stratiform events ($r=0.48$, $p\text{-value}=3,8 \cdot 10^{-6}$), we suggest that the wind plays a crucial role in influencing the removed of small-sized PS cells from the atmosphere. One hypothesis could be that small-sized raindrops experience reduced falling velocities during rain events characterized by high horizontal wind speeds. This could increase the probability of small-sized PS

cells being captured by raindrops, resulting in their removal from the atmosphere. A recent study (Luan et al., 2019) reported comparable findings, demonstrating that increased wind speed during rainfall can enhance the efficiency of scavenging particulate matter with a diameter of 2.5 μm (PM_{2.5}), especially during light rain intensities and longer durations. However, the mechanisms underlying this effect were not explained.

Like small-sized PS cells, the F_W of large-sized PS cells ($> 15 \mu\text{m}$) show weak correlation with the microphysical rainfall characteristics. During our study, larger-sized PS cells are mainly represented by PS cells rich in phycoerythrin (52% of the total F_W) (Fig. 2). A high F_W of large-sized of phycoerythrin-rich cells is reported only during specific events, classified as both convective and stratiform (G, I and K2) in February, which coincide with elevated levels of pollen as reported in previous studies (Devadas et al., 2018). Therefore, one possible explanation is that phycoerythrin-rich cells could be transported "attached" to pollen particles, as previously reported for the association between pollen particles and bacteria (Manirajan et al., 2018). Furthermore, the presence of large-sized PS cells rich in phycoerythrin is also observed during events originating from Saharan dust (SSW sector), suggesting that they may also be attached to dust particles, as previously reported for bacteria (Maki et al., 2017). Since organic and inorganic fragments such as pollen and dust particles have larger diameters than phycoerythrin-rich cells, the aggregation of these particles may increase their overall diameter and enhance gravitational settling (Mariraj Mohan, 2016; Pruppacher et al., 1998). Moreover, pollen is known to exhibit a strong auto-fluorescence (Pöhlker et al., 2013) and can emit in the same wavelength range as phycoerythrin-rich cells (Bozlee et al., 2005). This observation raises the possibility of misidentifying pollen particles as phycoerythrin-rich cells. Consequently, future monitoring efforts should consider using observational approaches to accurately distinguish phycoerythrin-rich cells from other atmospheric particles like pollen and mineral particles (dust and sea-salt).

4.2. Evidence of a long-distance transport of PS cells

Our findings also suggest evidence of a cloud contribution, in addition to the below-cloud scavenging, in explaining the temporal variability of the C_W of PS cells in rainwater. Indeed, a strong correlation is observed between the air masses transported over the Atlantic Ocean and the continents (SSW & ENE) and the C_W of PS cells suggesting that the atmospheric transport could be a main source of PS cells. This is confirmed by the biochemical composition of rainwater who contains tracers of distant emission sources (Xing et al., 2017; Violaki et al., 2022; Oduber et al., 2021). Long-range transport phenomena have been reported and show that marine ions can be transported from the Atlantic Ocean, while terrestrial ions can be emitted from Saharan dust events linked to the African continent. In our case, during events linked to oceanic origins (WNW), a marked correlation has been reported between the C_W of PS cells rich in chlorophyll (above 4 μm) and marine ions (Fig. 3b). Additionally, PS cells rich in phycoerythrin (2-15 μm) are correlated to the C_W of terrestrial ions during events associated with the African continent (SSW), whereas the C_W of small-sized cells (0-1 μm) are correlated with the

455 C_W of Cl^- during events characterized by an oceanic origin (Fig. 3d). Previous studies have also found
that PS cells are related to long-range transport, from both continental and oceanic origins (Dillon et
al., 2020; Lewandowska et al., 2017). This contribution of long-distance transport is also reinforced
by a strong correlation between the F_W and the C_W of medium to large-sized PS cells and the low-
altitude (below 2 km) movement of air masses, typically within the planetary boundary layer. This
460 layer has been identified as a significant source of biological aerosols, as it is in direct contact with the
surface where a substantial amount of living bioaerosols are found (Bryan et al., 2019). Additionally,
this layer also interacts with the overlying free troposphere, which is known for its capacity to
disperse aerosols regionally and globally through the long-range movement of air masses, driven by
high horizontal wind speeds (Šantl-Temkiv et al., 2022).

465 Chlorophyll-rich cells and phycoerythrin-rich cells, which are the dominant pigment classes during
the study period, are likely to include chlorophytes, cryptophytes, and cyanobacteria rich in
phycoerythrin based on their fluorescence and size characteristics measured in flow cytometry (Read
et al., 2014). Since their presence in clouds has already been shown with a specific role as cloud
condensation nuclei, initiating the formation of cloud droplets that can ultimately grow into raindrops
470 (Tesson and Šantl-Temkiv, 2018), other ecological roles of these PS cells could be suggested. Indeed,
previous studies have indicated that microalgae can survive and grow under cultural conditions after
being dispersed by rain (Tesson et al., 2016; Dillon et al., 2020). which suggests that these PS cells
have the potential to colonize new ecosystems. Therefore, conducting mesocosm studies and
employing genetic approaches such as omics methods would be beneficial in identifying the species
475 that survive after being disseminated by the rain. Subsequently, assessing whether these surviving
species can influence the long-term assemblages of phytoplankton communities in freshwater
ecosystems would provide valuable insights.

5. Conclusions

To our knowledge, studies reporting the analysis of the biochemical composition of rainwater coupled
480 to rain microphysical characteristics are rare, even non-existent. Based on our results, we suggest that
the scavenging of these photosynthetic cells vary according to their size and highlight distinct
mechanisms. We show that, during convective events, characterized by high rainfall intensity,
medium-sized photosynthetic cells (PS) are more efficiently scavenged, while smaller-sized are less
abundant in the rainwater, potentially due to their low below-cloud scavenging. In contrast, the
485 behavior of small-sized PS cells is primarily affected by horizontal wind speed, light rainfall
intensities, low raindrop velocity, and prolonged rain duration, which are common features of
stratiform rain events. Conversely, the deposition of large-sized cells is likely influenced by dry
deposition, regardless of the microphysical characteristics of the rainfall. The concentrations of PS
cells are also associated with various air mass origins, indicating multiple source emissions and
490 potential long-range transport. During high intensity rainfall events, we can expect important wet
atmospheric deposition of medium-sized PS cells which could influence the long-term assemblages of
phytoplankton communities via the contribution of new genotypes resistant to atmospheric conditions.
With the prospect of an increase in the intensity of rainfall in the coming years, we can wonder about

495 the effects of an increase in the contribution of PS atmospheric cells in the phytoplankton biodiversity
and the impacts on ecosystem functioning.

Author contributions

Fanny Noirmain: Writing - Original Draft, Conceptualization, Investigation, Visualization

500 **Jean-Luc Baray, Joel Van Baëlen & Delphine Latour:** Supervision, Review & Editing,
Conceptualization

Laurent Deguillaume: Review & Editing

Conflict of interest

The authors declare there is no conflict of interest.

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