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ABSTRACT

The coagulation of airborne particles on the surface of allergenic pollen grains is poorly described in the literature. However, particles deposited on the surface of pollen grains could have an effect on allergy and its symptoms. Observations of pollen surface alterations reported in the literature are either qualitative or are tainted by sampling artifacts that overestimate surface particulate pollution. Birch pollen grains (BPGs) were sampled in the atmosphere during pollination to quantitatively assess the extent of particulate deposition on allergenic pollen surfaces. Airborne PGs were collected with a cascade impactor ensuring no sampling artefact unlike pollen samplers usually used in this type of study. Counting, sizing and elemental analysis of particles adhering on pollen surfaces were performed by SEM/EDX. 68% of PGs did not carry any particle on their visible surface while the remaining 32% had between 1 to 16 particles on their surface. On average, polluted BPGs carried 2 ± 1 particles, representing an average surface coverage of 0.95%. We hypothesized that the main coagulation process was particle deposition by gravitational settling. Collision velocity calculations indicate that coagulation due to gravitational settling is prevalent under typical pollination atmospheric conditions. However, the effects of electric charges carried by pollen are largely unknown and may significantly alter coagulation rates. The presence of particle-polluted allergenic pollens appears to be common in the urban atmosphere, even under low air pollution conditions. The impact of chemical pollution of BPGs due to the presence of particulate matter (including soot particles) remains relatively unknown, both on the allergy and on the reproductive capacity of the tree.

1.INTRODUCTION

Birch pollen grains (BPGs) cause allergic reactions in more than 100 million people worldwide (Lavaud et al., 2013). An increasing trend in atmospheric BPG concentrations has been observed in several countries in relation to the relentless rise of carbon dioxide and the effects of climate change (Besancenot et al., 2019; Darbah et al., 2008; Frei, 1998; Hoebeke et al., 2017; Ziska et al., 2019). In addition, air pollution can aggravate allergy symptoms (Robichaud, 2021; Schiavoni et al., 2017). The allergic patient indeed never breathes BPGs alone; airborne BPGs are suspended in the urban atmosphere consisting of gaseous pollutants (NO_2, O_3, Volatile Organic Compounds), coarse, fine and ultrafine particles and other bioaerosols (spores and other potentially allergenic pollens). Taken separately, each of the constituents of the urban atmosphere can have aggravating effects on allergy but physical-chemical interactions also exist between gaseous and particulate atmospheric compounds. Alterations of BPGs due to atmospheric biochemical reactions with ozone or nitrogen dioxide have been highlighted (Sénéchal et al., 2015). Gaseous air pollutants modify the lipid and protein fraction of BPGs (Farah et al., 2021; Pereira et al., 2021; Visez et al., 2020). Not only pollen grains (PGs) can be modified by air pollution, but there are clear qualitative evidences that allergenic PGs can also act as carriers of pollutants, both adsorbed gases and coagulated particles (Okuyama et al., 2007; Zhu et al., 2018).

Particulate pollution of the surface of allergenic PGs is, unlike pollen-gas interactions, poorly assessed in the literature (Visez et al., 2020). Most of the observations of particles on the surface of PGs are qualitative reports that do not estimate the extent of the pollution of the surface of PGs by extrapollinic atmospheric particles. The available quantitative studies report particularly high pollution of PG surfaces. For

instance, Ribeiro et al. (2015) reported an average number of particles adhering to the pollen surface of 16 ± 12 (average ± standard deviation), 39 ± 32 and 4 ± 4 particles per PG for *Acer*, *Pinus* and *Platanus* pollen, respectively (Ribeiro et al., 2015). This study was conducted using a Hirst volumetric spore trap which is the most common method of sampling pollen on a moving slide, originally developed to count ambient pollen (Mullins & Emberlin, 1997). However, the use of a Hirst-type pollen sampler induces a sampling artifact with a significant overestimation of particle adhesion on PGs (Choël et al., 2020). In addition, to our knowledge, there are no quantitative data on the specific pollution of the surface of BPGs by atmospheric particulate matter (APM); only one study reported a high degree of particle agglomeration onto BPGs collected from two highly polluted urban areas (Behrendt et al., 1991).

Being able to sample windborne PGs while preserving their initial state is crucial to study how PM alters the physical, chemical and biological properties of PGs. PM pollution of PGs can only be adequately assessed if particle size-selective classification is employed when sampling airborne pollen. In such way, PGs can be efficiently trapped in a specific sampling stage (for coarse particles) different from smaller ambient particles (Levetin, 2004). Moreover, the sampling efficiency of airborne PGs is improved by the use of low nozzle velocity devices, such as impactors, compared to active sampling pollen trap, such as Hirst-type samplers (Choël et al., 2020; Ribeiro et al., 2015; Wittmaack et al., 2005). In addition, the sampling duration should be short enough (~a few hours) to avoid the pile-up of particles, especially in highly polluted environments (Wittmaack et al., 2005). The objective of this work is to document and reliably assess the size range, particle types and frequency of occurrence of particles adhered to airborne BPGs sampled under fairly

clean air conditions. Sampling of airborne PGs was carried out for the first time in the literature with a cascade impactor ensuring collection of PGs without piling up of fine particles on their surface in the collector during sampling (i.e., free from sampling artifact).

2.MATERIALS AND METHODS

Pollen sampling was carried out daily from 30 March to 7 April 2017 (start and end time: 10:00 am) on the roof of a building of Lille University campus (Lat. 50° 36' 29" N, Long. 3° 8' 25" E, Altitude 20 m a.s.l.) (Supplementary Figure S0) when weekly cumulative birch pollen concentrations peaked at 3022 grains·m^{-3} according to the French monitoring network (RNSA) in Lille metropolitan area (Figure 1). On 30 and 31 March 2017, daily birch pollen concentrations peaked at 986 and 1236 BPGs per m^3, respectively. Meteorological parameters were recorded by a weather station (Davis Vantage Pro) installed at the sampling site (Supplementary Figure S1).

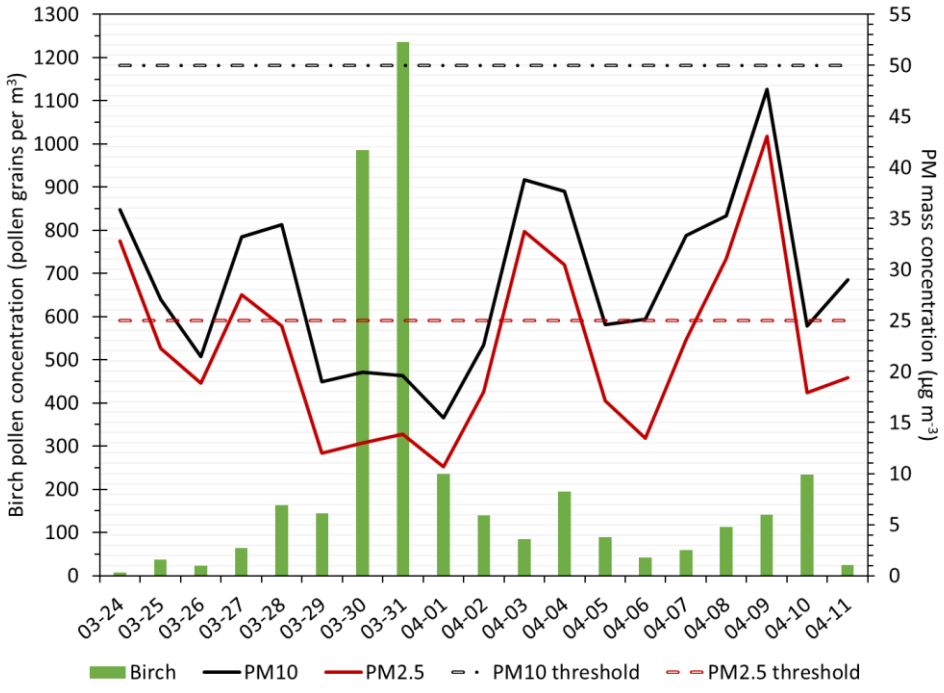

Figure 1. Daily concentrations of birch pollen grains in Lille metropolitan area (green bars) and ambient PM_{10} and $PM_{2.5}$ mass concentrations during pollen sampling (respectively black and red

curves). Threshold values recommended by the World Health Organization were reported in black dotted line for PM10 (50 $\mu g \cdot m^{-3}$) and in red dotted line for PM2.5 (25 $\mu g \cdot m^{-3}$).

No rain event occurred during the study period. Birch dehiscence was triggered by a temperature rise to 16°C combined with a drop of relative humidity on 30 March. The weekly average temperature, relative humidity, and wind speed were 11.9 ± 2.2 °C, 76.7 ± 5.1 % RH, 1.4 ± 0.5 $m \cdot s^{-1}$, respectively. Ambient PM_{10} and $PM_{2.5}$ concentrations were 26.3 ± 8.3 $\mu g \cdot m^{-3}$ and 19.3 ± 8.2 $\mu g \cdot m^{-3}$, respectively at the closest air quality monitoring station located 4 km away from the pollen sampling site (public data provided by ATMO Hauts-de-France). During the study period, PM_{10} concentrations never exceeded the WHO recommended threshold value, i.e. 50 $\mu g \cdot m^{-3}$ (24-hour mean). In contrast, $PM_{2.5}$ concentrations were over the WHO guideline of 25 $\mu g \cdot m^{-3}$ (24-hour mean) on 3 and 4 April 2017 (Figure 1). Airborne PGs were trapped on the first stage PM>10 µm of a 3-stage cascade impactor (PM10 impactor, Dekati Ltd.), i.e. impaction stage with particle size cut-off diameter of 10 µm at 50% collection efficiency valid for spherical particles with a mass density of 1 $g \cdot cm^{-3}$. The impactor was mounted on the top of a 4-meter-high mast (Supplementary Figure S2). Airborne PGs were impacted directly on a conductive carbon tape for electron microscopic examination. Bounce-off and re-entrainment of deposited PGs from the impaction surfaces can be minimized by using adhesive collection substrates (Wittmaack et al., 2005).

A number of 236 PGs were observed directly on the collection substrate without metal coating using a low-vacuum SEM (JEOL JSM 7800F LV) operated at a working pressure of 100 Pa in the specimen chamber. Visual detection of BPGs was carried out by low magnification SEM inspection of the whole area of daily collection substrates (circular surface with a diameter of 25 mm) (Figure 2).

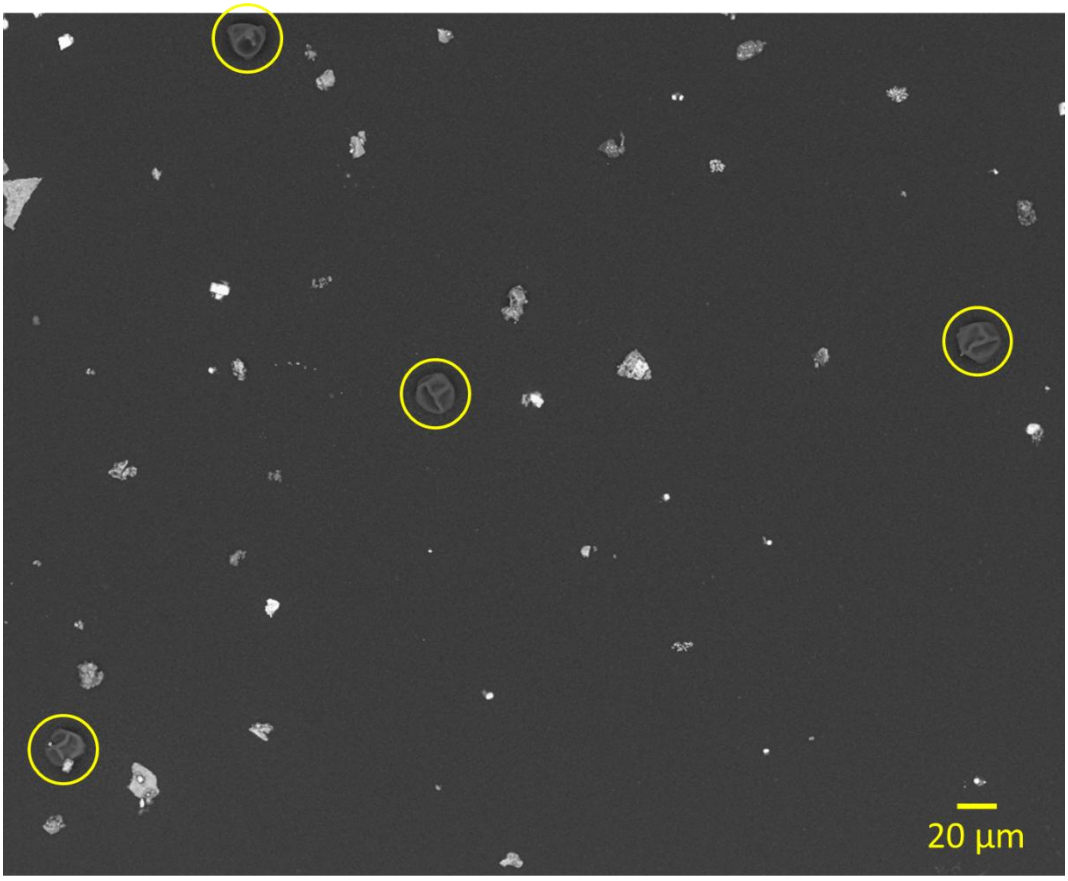

Figure 2. Example of SEM image of birch pollen grains (circled) and atmospheric particulate matter deposited on PM>10 µm impaction stage. The pollen grain on the bottom left carries atmospheric particles.

As the impaction surface was static in the case of our PM10 impactor, large particles might pile up on top of pollen grains during sampling. The probability of having a particle pile-up increases with the sampling duration. A typical view of the substrate loading in the case of our daily sampling is presented in Figure 2. As can be seen, after 24-hour sampling on $PM_{>10}$ impaction stage, the particulate load on the collection substrate was low, with BPGs well-separated from the other collected particles, ensuring a minimized sampling artifact (Choël et al. 2020).

BPGs were detected by using backscattered electron (BSE) imaging. Because BSE yield depends on the atomic number of the atoms causing the backscattering, BSE images are sensitive to the elemental make-up of particles. BPGs are predominantly

made up of light elements such as the carbon substrate. As a result, BPGs and the substrate appear with similar grey levels. To confirm the presence of a BPG and to examine particulate adhesion, each BPG was magnified and secondary electron (SE) images sensitive to topography were taken. Counting and sizing of particles adhering to pollen surfaces were determined from BSE images using FIJI/ImageJ software. X-ray elemental analysis of particles attached to PGs was carried out with an energy-dispersive X-ray (EDX) detector (SDD 80 mm^2, Oxford Instruments Inc.) at an accelerating voltage of 15 kV. Note that BPGs appear deflated as a result of shrinkage due to low-vacuum which enables EDX analysis of micrometer-sized particles down to 0.2 µm (Choël et al., 2010).

3.RESULTS

A total of 236 BPGs were individually imaged by low-vacuum SEM/EDX and a total of 205 particles were observed on birch pollen surfaces. 68% of PGs did not carry any particle on their visible surface. The remaining 32% of PGs had between 1 and 16 particles adhered to their surface. On average, polluted PGs had 2 ± 1 particles on their surface. Sampling artifact was assessed to 2.2 ± 2.4 particles per PG with a conventional Hirst-type sampler under similar PM10 and PM2.5 concentrations (Choël et al., 2020). The low number of particles observed on birch pollen (2 ± 1 particles per BPG) confirms the need to use an impactor because the sampling artifact of an HTS is of the same order of magnitude.

Particles deposited on BPGs had an average diameter of 1.5 ± 2.0 µm (average ± standard deviation) and ranged in size from 0.2 to 11.8 µm. Polluted PGs had particles covering an average of 0.95% of their surface. Particles lying on the substrate had an average diameter of 5.6 ± 4.0 µm with sizes varying between 0.3 and 21.9 µm (total number of particles observed on the substrate=309). Both the average

diameter and the size range were larger on the collection substrate compared to PG surface, indicating a good size differentiation between PGs and fine particles in suspension using $PM_{>10}$ impaction sampling device. Two other types of PGs, namely willow (*Salix* L.) and hornbeam (*Carpinus betulus*), were also observed with particles adhering on their surface (Supplementary figures S3 and S4).

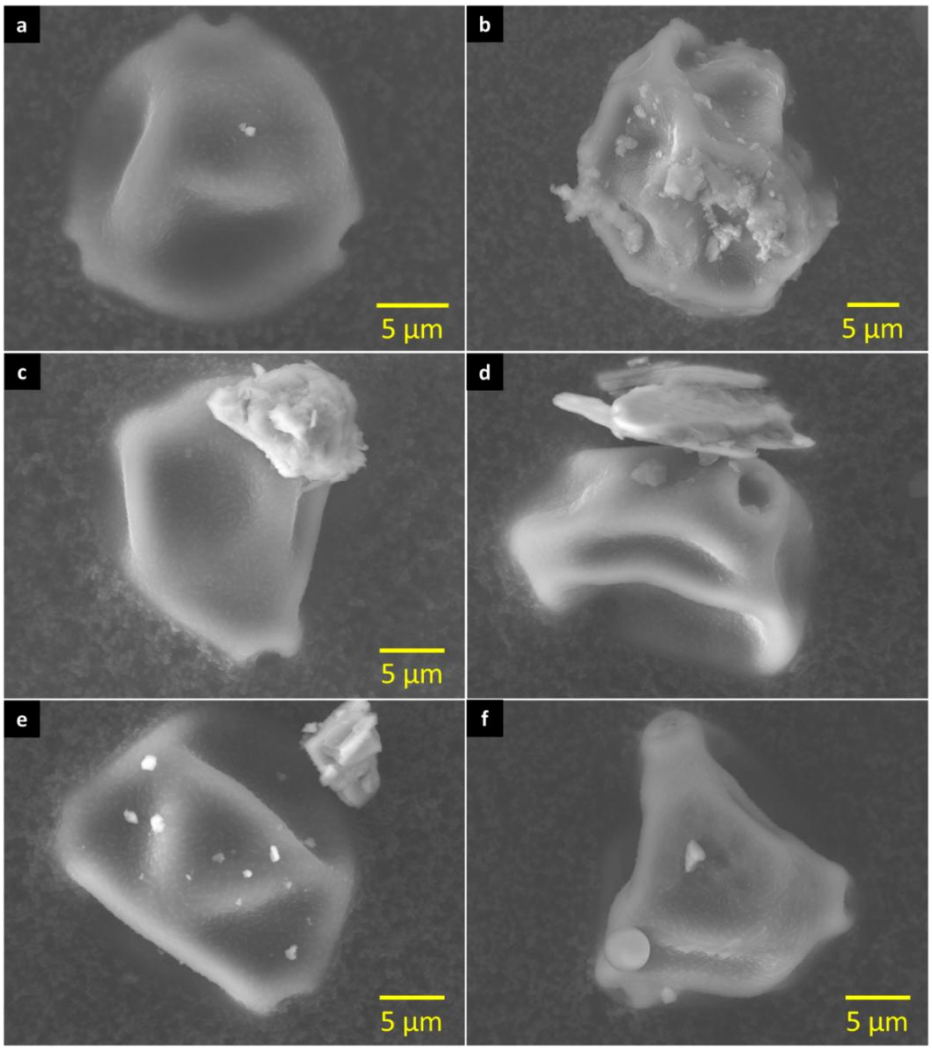

Figure 3. SEM images of polluted birch pollen grains carrying: (a) 1 µm-sized calcium silicate particle; (b) aluminosilicate particles; (c) a large soil dust particle (plagioclase Al-Si-Ca-Fe-rich); (d) a large phyllosilicate particle, (e) one supermicron and a dozen submicron dolomite particles; (f) a spherical coal fly ash particle (O-Al-Si-rich diam 3.5 µm), a calcite particle (up) and a gypsum particle (down).

The nature of particles adhered to BPGs were as follow (Figure 3 and Figure 4): soil dust (calcite, quartz, orthoclase (potassium feldspath), plagioclase, olivine, clay

minerals (illite, smectite, mica)), sea-salts, metal-rich (Fe-rich, Cu-rich, Ni-rich, Zn-rich, Ba-rich) particles, small and large soot aggregates consisting of clusters of nanospheres with a diameter of 40 to 90 nm.

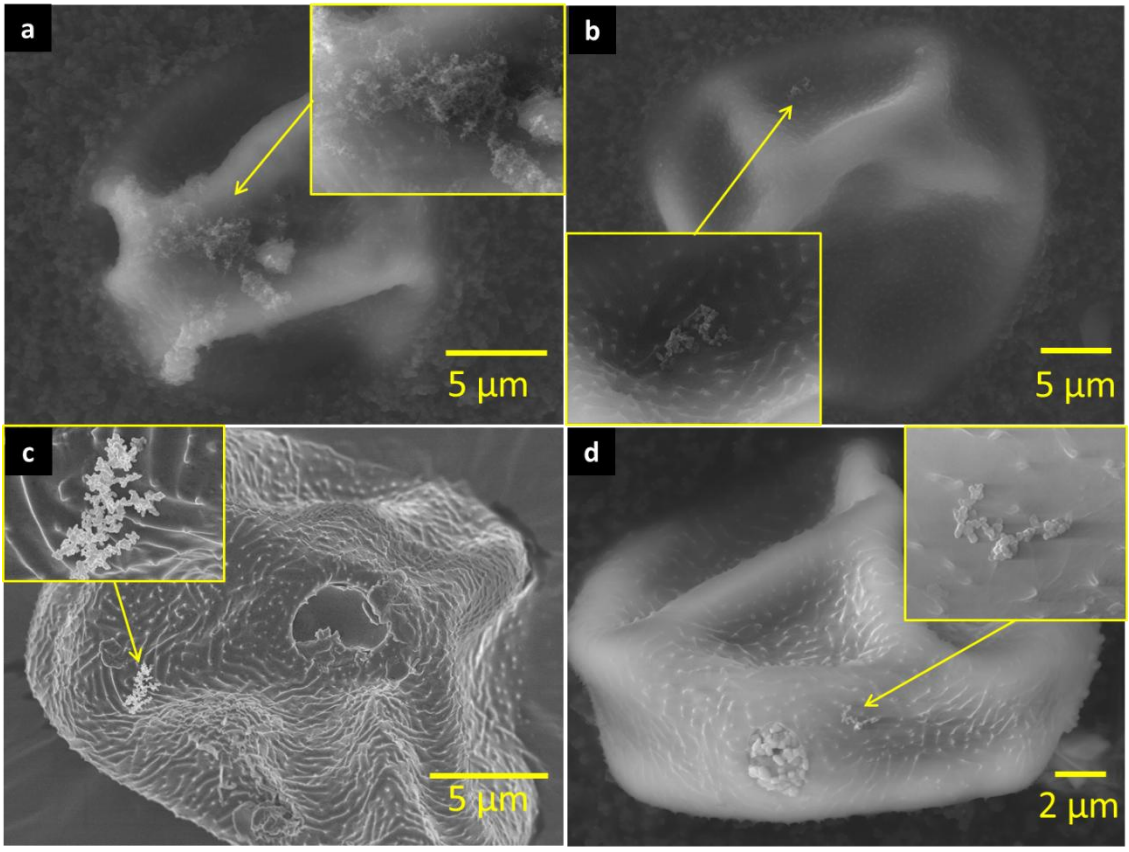

Figure 4. SEM images of polluted birch pollen grains: (a) covered with a large chain-like soot aggregate of nanospheres (diameter 40-50 nm); (b-c) carrying a micrometer-sized chain-like soot aggregate of nanospheres (diameter 80-90 nm); (d) with a soot aggregate (area equivalent diameter 1.1 µm) and recrystallized sea-salts.

4.DISCUSSION

Our atmospheric samples collected under conditions of moderate particulate air pollution reveals that pollution of the surface of pollen grains (PGs) by atmospheric particles is a common phenomenon: out of a total of 236 birch pollen grains (BPGs) examined, 32% of BPGs carried an average of 2 ± 1 particles (205 particles deposited on 75 BPGs). On a global average during our sampling, BPGs carried 0.86 particles (205 particles observed particles for 236 BPGs). The fraction of the pollen surface

202 covered by particles is however very low, less than 1%, which contrasts with the
203 higher fractions previously reported in the literature (Behrendt et al., 1991; Ribeiro et
204 al., 2015). For example, the area covered by particles on pine and maple pollen
205 collected from the atmosphere with a Hirst trap was 5.7% and 7.5%, respectively
206 (Ribeiro et al., 2015). A very high degree of agglomeration was also observed for BPG
207 collected with a high volume sampler in highly polluted urban areas (Behrendt et al.,
208 1991). We assume that the high degrees of agglomeration presented in these previous
209 works may be subject to a sampling artefact involving an overestimation of the
210 number of particles per PG. Hirst-type samplers are not suitable for the assessment
211 of particulate pollution on the surface of PGs due to a too high impaction velocity
212 which does not allow the discrimination of pollen and PM10 during sampling (Choël
213 et al., 2020). The use of a PM10 impactor avoids this sampling artefact and explains,
214 at least in part, the lower particulate pollution rate obtained in this study (Choël et
215 al., 2020). Another explanation is that the air quality was relatively good in our study
216 (PM10 = 26.3 ± 8.3 $\mu g \cdot m^{-3}$), which reduces the probability of coagulation of
217 atmospheric particles on the surface of PGs. One of the limitations of our study is the
218 low number of PGs observed. Manual grain-by-grain observation by electron
219 microscopy is indeed time-consuming and unsuitable for samples of several thousand
220 grains but it is the only way to reliably assess the presence of particles on PGs. In
221 addition, some particles are difficult to identify, in particular due to the presence of
222 sub-pollen particles on the surface of BPGs (Ruggiero & Bedini, 2018, 2020). It
223 should also be remembered that SEM analysis only allows to observe the visible
224 pollen surface; particles stuck to the pollen surface on the side of the collection
225 substrate (hidden surface) are obviously not observable.

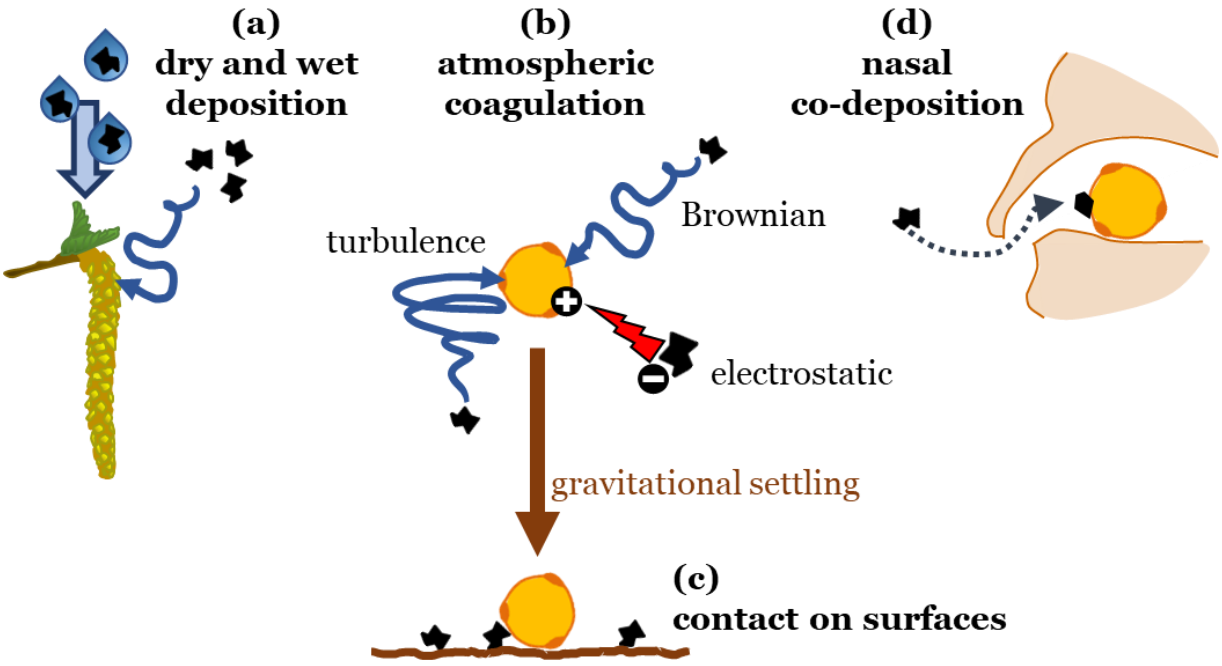

Figure 5. Proposed mechanisms for coagulation of atmospheric particles on the surface of pollen grains. The yellow balls represent birch pollen grains. The black pictograms symbolize atmospheric particles.

The pollution rate of BPGs obtained in our sampling can be explained by several mechanisms of APM deposition (Figure 5) without the possibility to assign their respective relative contributions: dry and wet deposition on catkins before pollination (a), coagulation in the atmosphere (b), contact after deposition on a surface polluted by particles followed by resuspension of the PG (c). Particulate pollution on catkins and resuspension of PGs need to be specifically studied as these mechanisms are poorly documented (Visez et al., 2020).

On the other hand, it is possible to estimate by calculation the contribution of coagulation of particles with PGs in the atmosphere (Figure 5 (b)) ; particles coagulation is described by the rate of collision J_{12} (particles.s^{-1}) between two populations of particles at concentrations N_1 and N_2 (equation 1, Table) (Seinfeld & Pandis, 2016). Equations 1 to 4 (Table) describe respectively Brownian coagulation, gravitational coagulation and coagulation in turbulent flow (García-Nieto, 2006;

Seinfeld & Pandis, 2016). The coagulation rates could also be influenced by the electrical charges of particles (Harrison & Carslaw, 2003) ; however, the lack of data on the distribution of electrical charges in our sampling conditions does not allow to estimate this specific contribution (enhancement, no effect or reduction) on the deposition rates of APM on BPGs.

Table 1. Equations for the rate of collision J_{12} between particles 1 and 2 (eq. 1) and coagulation rate coefficients K_{12} for Brownian coagulation (eq. 2), gravitational coagulation (eq. 3) and coagulation in turbulent flow (eq. 4) (García-Nieto, 2006; Seinfeld & Pandis, 2016).

Equation 1 **Rate of collision**	$J_{12} = K_{12}N_1N_2$	N particle concentration (#.m^{-3}) K_{12} coagulation rate coefficients
Equation 2 **Brownian coagulation**	$K_{12}^{BG} = \frac{2kT}{3\mu}(D_{P1} + D_{P2})(\frac{C_{C1}}{D_{P1}}+\frac{C_{c2}}{D_{P2}})$	k Boltzmann constant; μ viscosity of air (20°C, 1.825×10^{-5} kg·m^{-1}·s^{-1}); T temperature (K); D_P diameter of particles (m); C_c Cunningham correction factor
Equation 3 **Gravitational coagulation**	$K_{12}^{GC} = \frac{\pi}{4}(D_{P1} + D_{P2})^2(\vartheta_{t1} - \vartheta_{t2})E(D_{P1,}D_{P2})$	ϑ_t terminal settling velocity (BPGs = 0.012 m·s^{-1} ((Sofiev et al., 2006)); $E(D_{P1,}D_{P2})$ collision efficiency (=1 in this work)
Equation 4 **Coagulation in turbulent flow**	$K_{12}^{TF} = \left(\frac{\pi^2\varepsilon_k}{120\upsilon}\right)^{1/2}(D_{P1} + D_{P2})^3$	ε_k rate of dissipation of kinetic energy per unit mass (10 s^{-1}); υ kinematic viscosity of fluid = (15.06×10^{-6} m²·s)

Calculations of K_{12} for the three types of coagulation described in Table were performed for a particle deposition on BPG (21 μm in diameter) from a monomodal population of 2 μm diameter particles at a concentration of 5 particules·cm^{-3},

considering a terminal settling velocity of 100 cm·h^{-1} for particles (1.2 cm·s^{-1} for BPG) and no rebounds after collision. This computational estimate shows that K_{12} is of the same order of magnitude for gravitational coagulation ($K_{12}^{GC} \approx$ 4.8×10^{-12} m^6·s^{-1}) as for turbulence ($K_{12}^{TF} \approx$ 1.6×10^{-12} m^6·s^{-1}) while K_{12}^{BG} related to Brownian motion is largely negligible ($K_{12}^{BG} \approx$ 2×10^{-15} m^6·s^{-1}). Equation 3 tends to predict very high coagulation with the finer particles, as the difference in settling velocity is greater and the concentrations of the fine particles are several thousands to tens of thousands of particles per cm^3. However, the efficiency of collision E_{12}, which depends on the size of the particles involved, decreases sharply as the diameter of particle 2 tends towards zero (Seinfeld & Pandis, 2016). This prediction is consistent with our observations where relatively few very fine particles (< 500 nm) were detected on the surface of BPGs.

Under typical pollination weather conditions (absence of turbulence, calm weather and low wind), and without taking into account electrostatic interactions between particles, the main coagulation process would be the deposition of particles by gravitational settling. By assuming a concentration of 2-µm-sized particles N_2 in the order of 5 particles.cm^{-3} and a gravitational settling time of one hour (drop of 43 m), we calculate 0.09 particles deposited per PG (J_{12}=2.4×10^{-5} particles·s^{-1}). For pollen transported over long distances, while some PGs have been observed at considerable altitudes of 2 km (Damialis et al., 2017), it is conceivable that gravitational settling alone explains the levels of particulate contamination of PGs observed in our samples (on average 0.86 particles per BPG). If the hypothesis of coagulation primarily by gravitational settling is correct, then the presence of particles on the surface of BPGs could be a marker of atmospheric transport of pollen over long distances.

279 Our coagulation calculations shed interesting light on the hypothesis of increased
280 COVID-19 propagation by virus transport on the surface of PGs (Damialis et al., 2021;
281 Dbouk & Drikakis, 2021). The thousands of droplets emitted by exhalation, and
282 potentially infected by a virus, are in the micrometre to 100 μm range, with a
283 maximum around 50 to 150 μm (Xie et al., 2009). Our estimates show that the
284 encounter between a PG and a coarse particle of about 100 μm will be very unlikely
285 for contact times of a few minutes. For fine particles (2 μm) exhaled at a
286 concentration of 150,000 particles·m^{-3} in contact with a high concentration of BPGs
287 (1,000 BPGs·m^{-3}), the calculated deposition rate of 2-μm-sized particles on pollen is
288 0.06 particles per day (Shao et al., 2021). In light of these estimates of the
289 coagulation rate and the low number of particles observed on the surface of pollen in
290 our atmospheric samples, the transport by a PG of a virus expectorated by a human
291 being is most likely a rare event. If pollen is involved in the aggravation of COVID
292 infection (Gilles et al., 2021; Khaiwal et al., 2021), we suggest that co-exposure to
293 expectorated droplets and PGs is more likely involved than virus adhesion to the
294 pollen surface. More precise modelling work is needed; our estimates by calculating
295 the coagulation rate of particles on pollen are only intended to give orders of
296 magnitude of the coagulation phenomena involved.

297 The observation of several PGs with soot suggests that combustion-emitted particles
298 may have a particular affinity with pollen. It has been suggested that electrostatic
299 interactions may promote affinity between combustion particles and bioaerosols
300 (Roos & Dutertre-Laduree, 1990). However, the number of BPGs with soot is too
301 small to provide a reliable estimate of occurrence of soot sticking on BPGs in the
302 urban atmosphere. Furthermore, the sampling was conducted on the roof of a
303 building (20 m high); sampling at 1.5 m above the ground could promote soot-pollen

304 coagulation because, on the one hand, airborne particle concentrations can be as high
305 as 10^6 to 10^7 particles per cm^3 (Seinfeld & Pandis, 2016) and, on the other hand,
306 pollen concentrations can be higher (Charalampopoulos et al., 2021). Further
307 sampling, especially at street level, is therefore necessary to better assess the fraction
308 of PGs polluted by soot, especially as a large body of literature highlights the
309 aggravating effects on allergy of co-exposure to pollen and soot (Chen et al., 2020;
310 Nel et al., 1998; Shiraiwa et al., 2012). Unlike nasal co-deposition (Figure 5), the
311 pollen and soot (or pollen and particles) mixture resulting from atmospheric
312 coagulation may have time (up to several days) to undergo atmospheric aging.
313 Chemical reactivity at the particle-pollen contact surface cannot be excluded. Very
314 little information is available on this multiphase pollen chemistry; for example, it has
315 been shown that in the presence of moisture, allergens can diffuse from the PG to
316 soot particles deposited on its surface (Knox et al., 1997; Ormstad et al., 1998).

317 Birch pollen is never breathed alone; each inhalation introduces gaseous pollutants,
318 particles, other allergenic pollens, and other bioaerosols (viruses, bacteria, fungus
319 spores or other non-allergenic pollens) into the respiratory system. A central question
320 will be whether the level of particle deposition observed in this study on airborne
321 BPGs (0.86 particles per PG) is significant in comparison with the effects of co-
322 exposure to pollen and APM during breathing (Figure 5d). First, it should be noted
323 that the effects of these two mechanisms are cumulative; co-deposition also occurs on
324 PGs that have already been subjected to coagulation of atmospheric particles. In
325 addition, and as previously mentioned, atmospheric aging involving at least
326 biochemical reactivity and diffusion of allergens may occur after coagulation during
327 atmospheric transport of BPGs. Our study alone cannot determine the potential
328 impacts of pollen surface particle pollution on allergy. However, our atmospheric

observations allow to determine, for the first time without sampling artifacts, the particle pollution state of the surface of inhaled pollen, which may prompt future *in vitro* studies on the effects of the particle-pollen cocktail to use realistic particle contamination of the pollen surfaces (on the order of 1%).

Finally, it should be mentioned that, beyond the health effects, the adhesion of particles to the pollen surface could influence the reproductive capacity of pollen. Some studies show a negative effect of metallic nanoparticles (Pd, Fe, ZnO) and carbon particles (fullerene, graphene) on pollen reproductive capacity (Aoyagi & Ugwu, 2011; Carniel et al., 2018; Dutta Gupta et al., 2019; Speranza et al., 2010, 2013; Yoshihara et al., 2021). However, these studies use particularly high particle-to-pollen ratios and chemical compositions that are not very representative of real atmospheric particles. It was also shown that APM does not interfere with the germination of *Cichorium intybus* pollen along roadsides (Jaconis et al., 2017). Moreover, if the hypothesis of greater pollution of the pollen surface by particles during long-distance transport is correct, it appears that the reduction of germination by particle adhesion is probably insignificant because the probability of reproduction between two trees located at a long distance is itself particularly low, if not zero (Proctor et al., 1996).

5.CONCLUSION

An average pollen surface contamination of 2 ± 1 particles per BPG was obtained for 38% of BPGs sampled under moderate air pollution conditions (average PM10 = 26.3 $\mu g \cdot m^{-3}$). This value was not known in the literature because previous estimates were possibly subject to sampling artifacts. Determination of this particulate contamination level is important for future studies on germination or allergy as it

353 provides an estimate of a realistic dose-response effect of APM coagulated on pollen
354 surface.

355 BPGs are small (21 μm in diameter) and larger pollens could have higher APM
356 deposition rates. Gravitational settling is indeed suspected be the predominant
357 mechanism of coagulation in the atmosphere. Very little is known about pollen
358 pollution by APM on the plant, about electrostatic interactions between particles and
359 also about pollen resuspension after deposition on a surface. APM coagulation on
360 PGs could be particularly enhanced during specific atmospheric events, such as
361 thunderstorms, which can, for example, alter the electrical charges of APM and PGs,
362 increase turbulence conditions, and generate large amplitude vertical transport of
363 PGs.

364 In our current atmosphere, on a PG surface of only 1,400 μm², traces of particulate
365 pollution were found on one out of three PGs. The types of particles deposited on the
366 PGs reflect the influence of human activities intensively emitting particles:
367 desertification, urban sprawl, forest fires (accentuated by climate change), intensive
368 combustion of fossil fuels and excessive industrialization.

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