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# 1           **Quantitative Assessment of Coagulation of Atmospheric**

## 2                   **Particles onto Airborne Birch Pollen Grains**

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19   sampling, pollen allergy.

20 **ABSTRACT**

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

43

44

## 45 **1.INTRODUCTION**

46 Birch pollen grains (BPGs) cause allergic reactions in more than 100 million people  
47 worldwide (Lavaud et al., 2013). An increasing trend in atmospheric BPG  
48 concentrations has been observed in several countries in relation to the relentless rise  
49 of carbon dioxide and the effects of climate change (Besancenot et al., 2019; Darbah  
50 et al., 2008; Frei, 1998; Hoebeke et al., 2017; Ziska et al., 2019). In addition, air  
51 pollution can aggravate allergy symptoms (Robichaud, 2021; Schiavoni et al., 2017).  
52 The allergic patient indeed never breathes BPGs alone; airborne BPGs are suspended  
53 in the urban atmosphere consisting of gaseous pollutants (NO<sub>2</sub>, O<sub>3</sub>, Volatile Organic  
54 Compounds), coarse, fine and ultrafine particles and other bioaerosols (spores and  
55 other potentially allergenic pollens). Taken separately, each of the constituents of the  
56 urban atmosphere can have aggravating effects on allergy but physical-chemical  
57 interactions also exist between gaseous and particulate atmospheric compounds.  
58 Alterations of BPGs due to atmospheric biochemical reactions with ozone or nitrogen  
59 dioxide have been highlighted (Sénéchal et al., 2015). Gaseous air pollutants modify  
60 the lipid and protein fraction of BPGs (Farah et al., 2021; Pereira et al., 2021; Visez et  
61 al., 2020). Not only pollen grains (PGs) can be modified by air pollution, but there are  
62 clear qualitative evidences that allergenic PGs can also act as carriers of pollutants,  
63 both adsorbed gases and coagulated particles (Okuyama et al., 2007; Zhu et al.,  
64 2018).

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

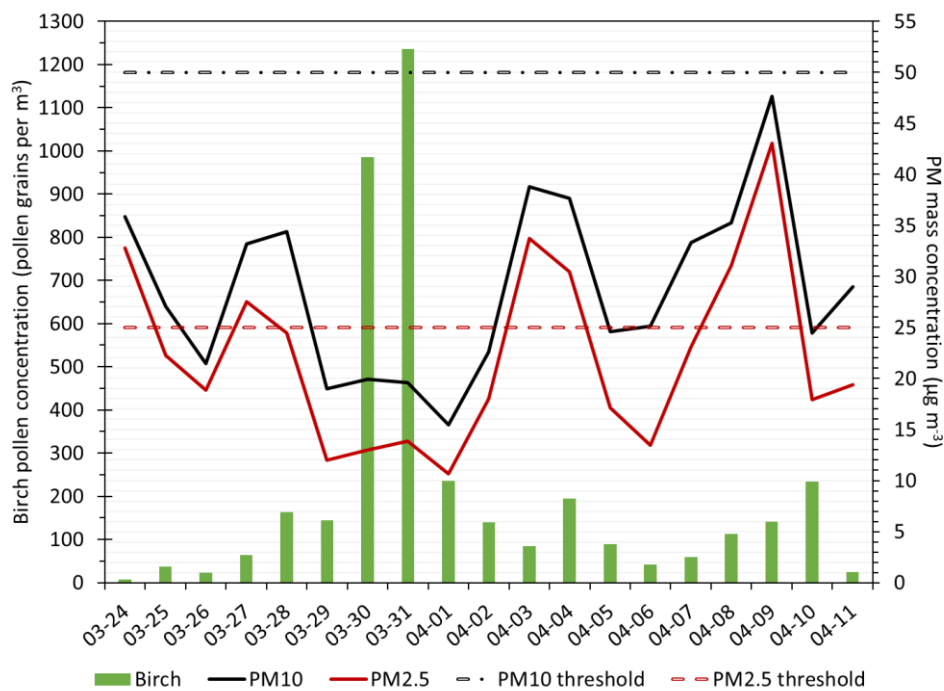
70 instance, Ribeiro et al. (2015) reported an average number of particles adhering to  
71 the pollen surface of  $16 \pm 12$  (average  $\pm$  standard deviation),  $39 \pm 32$  and  $4 \pm 4$   
72 particles per PG for *Acer*, *Pinus* and *Platanus* pollen, respectively (Ribeiro et al.,  
73 2015). This study was conducted using a Hirst volumetric spore trap which is the  
74 most common method of sampling pollen on a moving slide, originally developed to  
75 count ambient pollen (Mullins & Emberlin, 1997). However, the use of a Hirst-type  
76 pollen sampler induces a sampling artifact with a significant overestimation of  
77 particle adhesion on PGs (Choël et al., 2020). In addition, to our knowledge, there are  
78 no quantitative data on the specific pollution of the surface of BPGs by atmospheric  
79 particulate matter (APM); only one study reported a high degree of particle  
80 agglomeration onto BPGs collected from two highly polluted urban areas (Behrendt  
81 et al., 1991).

82 Being able to sample windborne PGs while preserving their initial state is crucial to  
83 study how PM alters the physical, chemical and biological properties of PGs. PM  
84 pollution of PGs can only be adequately assessed if particle size-selective  
85 classification is employed when sampling airborne pollen. In such way, PGs can be  
86 efficiently trapped in a specific sampling stage (for coarse particles) different from  
87 smaller ambient particles (Levetin, 2004). Moreover, the sampling efficiency of  
88 airborne PGs is improved by the use of low nozzle velocity devices, such as impactors,  
89 compared to active sampling pollen trap, such as Hirst-type samplers (Choël et al.,  
90 2020; Ribeiro et al., 2015; Wittmaack et al., 2005). In addition, the sampling  
91 duration should be short enough (~a few hours) to avoid the pile-up of particles,  
92 especially in highly polluted environments (Wittmaack et al., 2005). The objective of  
93 this work is to document and reliably assess the size range, particle types and  
94 frequency of occurrence of particles adhered to airborne BPGs sampled under fairly

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

## 99 2. MATERIALS AND METHODS

100 Pollen sampling was carried out daily from 30 March to 7 April 2017 (start and end  
 101 time: 10:00 am) on the roof of a building of Lille University campus (Lat. 50° 36' 29"  
 102 N, Long. 3° 8' 25" E, Altitude 20 m a.s.l.) (Supplementary Figure S0) when weekly  
 103 cumulative birch pollen concentrations peaked at 3022 grains·m<sup>-3</sup> according to the  
 104 French monitoring network (RNSA) in Lille metropolitan area (Figure 1). On 30 and  
 105 31 March 2017, daily birch pollen concentrations peaked at 986 and 1236 BPGs per  
 106 m<sup>3</sup>, respectively. Meteorological parameters were recorded by a weather station  
 107 (Davis Vantage Pro) installed at the sampling site (Supplementary Figure S1).



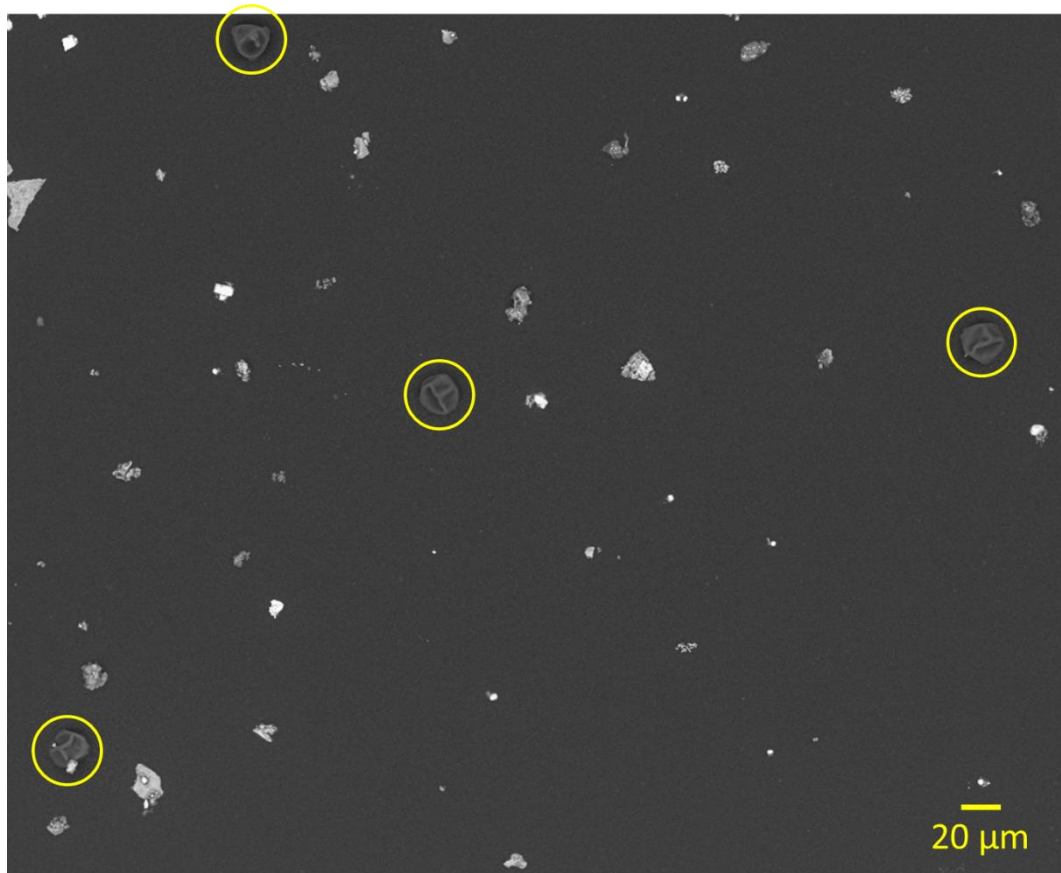
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109 Figure 1. Daily concentrations of birch pollen grains in Lille metropolitan area (green bars) and  
 110 ambient PM<sub>10</sub> and PM<sub>2.5</sub> mass concentrations during pollen sampling (respectively black and red

111 curves). Threshold values recommended by the World Health Organization were reported in black  
112 dotted line for PM<sub>10</sub> (50 µg·m<sup>-3</sup>) and in red dotted line for PM<sub>2.5</sub> (25 µg·m<sup>-3</sup>).

113 No rain event occurred during the study period. Birch dehiscence was triggered by a  
114 temperature rise to 16°C combined with a drop of relative humidity on 30 March. The  
115 weekly average temperature, relative humidity, and wind speed were 11.9 ± 2.2 °C,  
116 76.7 ± 5.1 % RH, 1.4 ± 0.5 m·s<sup>-1</sup>, respectively. Ambient PM<sub>10</sub> and PM<sub>2.5</sub> concentrations  
117 were 26.3 ± 8.3 µg·m<sup>-3</sup> and 19.3 ± 8.2 µg·m<sup>-3</sup>, respectively at the closest air quality  
118 monitoring station located 4 km away from the pollen sampling site (public data  
119 provided by ATMO Hauts-de-France). During the study period, PM<sub>10</sub> concentrations  
120 never exceeded the WHO recommended threshold value, i.e. 50 µg·m<sup>-3</sup> (24-hour  
121 mean). In contrast, PM<sub>2.5</sub> concentrations were over the WHO guideline of 25 µg·m<sup>-3</sup>  
122 (24-hour mean) on 3 and 4 April 2017 (Figure 1). Airborne PGs were trapped on the  
123 first stage PM<sub>>10</sub> µm of a 3-stage cascade impactor (PM<sub>10</sub> impactor, Dekati Ltd.), i.e.  
124 impaction stage with particle size cut-off diameter of 10 µm at 50% collection  
125 efficiency valid for spherical particles with a mass density of 1 g·cm<sup>-3</sup>. The impactor  
126 was mounted on the top of a 4-meter-high mast (Supplementary Figure S2). Airborne  
127 PGs were impacted directly on a conductive carbon tape for electron microscopic  
128 examination. Bounce-off and re-entrainment of deposited PGs from the impaction  
129 surfaces can be minimized by using adhesive collection substrates (Wittmaack et al.,  
130 2005).

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



136

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

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

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



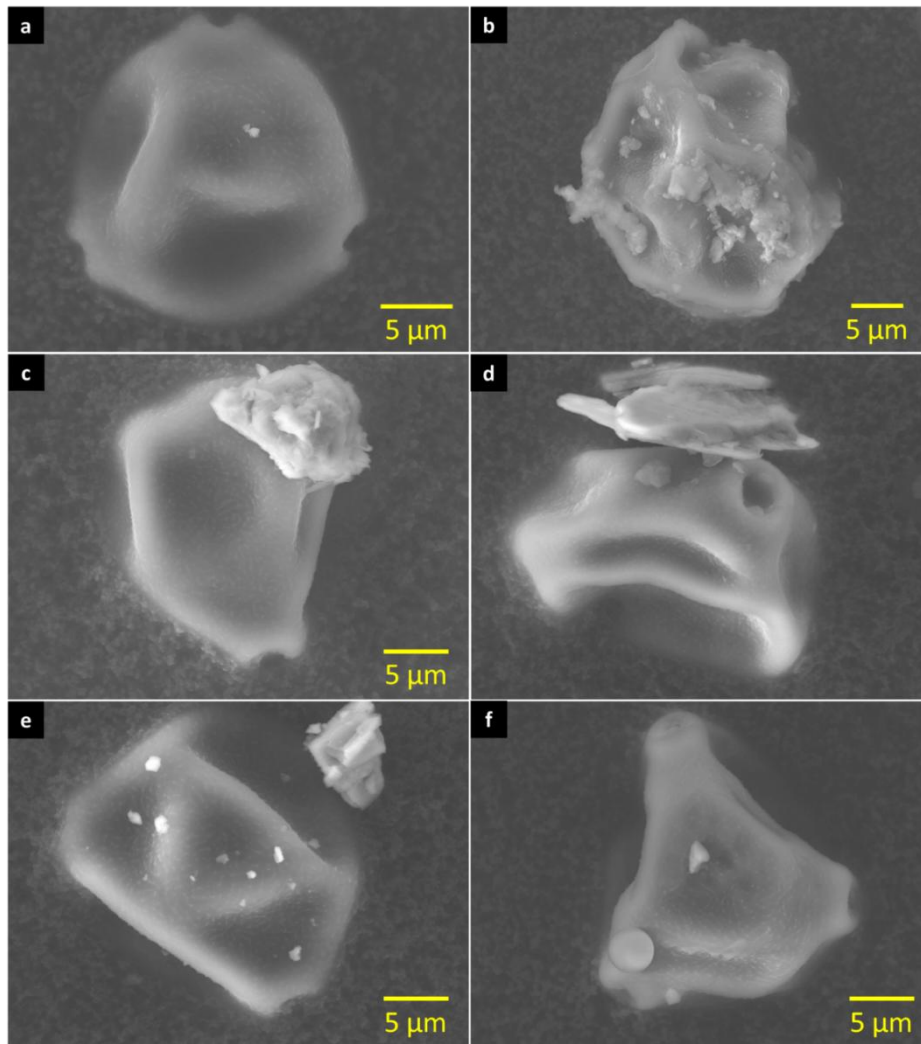
150 made up of light elements such as the carbon substrate. As a result, BPGs and the  
151 substrate appear with similar grey levels. To confirm the presence of a BPG and to  
152 examine particulate adhesion, each BPG was magnified and secondary electron (SE)  
153 images sensitive to topography were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

### 160 **3.RESULTS**

161 A total of 236 BPGs were individually imaged by low-vacuum SEM/EDX and a total  
162 of 205 particles were observed on birch pollen surfaces. 68% of PGs did not carry any  
163 particle on their visible surface. The remaining 32% of PGs had between 1 and 16  
164 particles adhered to their surface. On average, polluted PGs had  $2 \pm 1$  particles on  
165 their surface. Sampling artifact was assessed to  $2.2 \pm 2.4$  particles per PG with a  
166 conventional Hirst-type sampler under similar PM<sub>10</sub> and PM<sub>2.5</sub> concentrations  
167 (Choël et al., 2020). The low number of particles observed on birch pollen ( $2 \pm 1$   
168 particles per BPG) confirms the need to use an impactor because the sampling  
169 artifact of an HTS is of the same order of magnitude.

170 Particles deposited on BPGs had an average diameter of  $1.5 \pm 2.0$   $\mu\text{m}$  (average  $\pm$   
171 standard deviation) and ranged in size from 0.2 to 11.8  $\mu\text{m}$ . Polluted PGs had  
172 particles covering an average of 0.95% of their surface. Particles lying on the  
173 substrate had an average diameter of  $5.6 \pm 4.0$   $\mu\text{m}$  with sizes varying between 0.3 and  
174 21.9  $\mu\text{m}$  (total number of particles observed on the substrate=309). Both the average

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

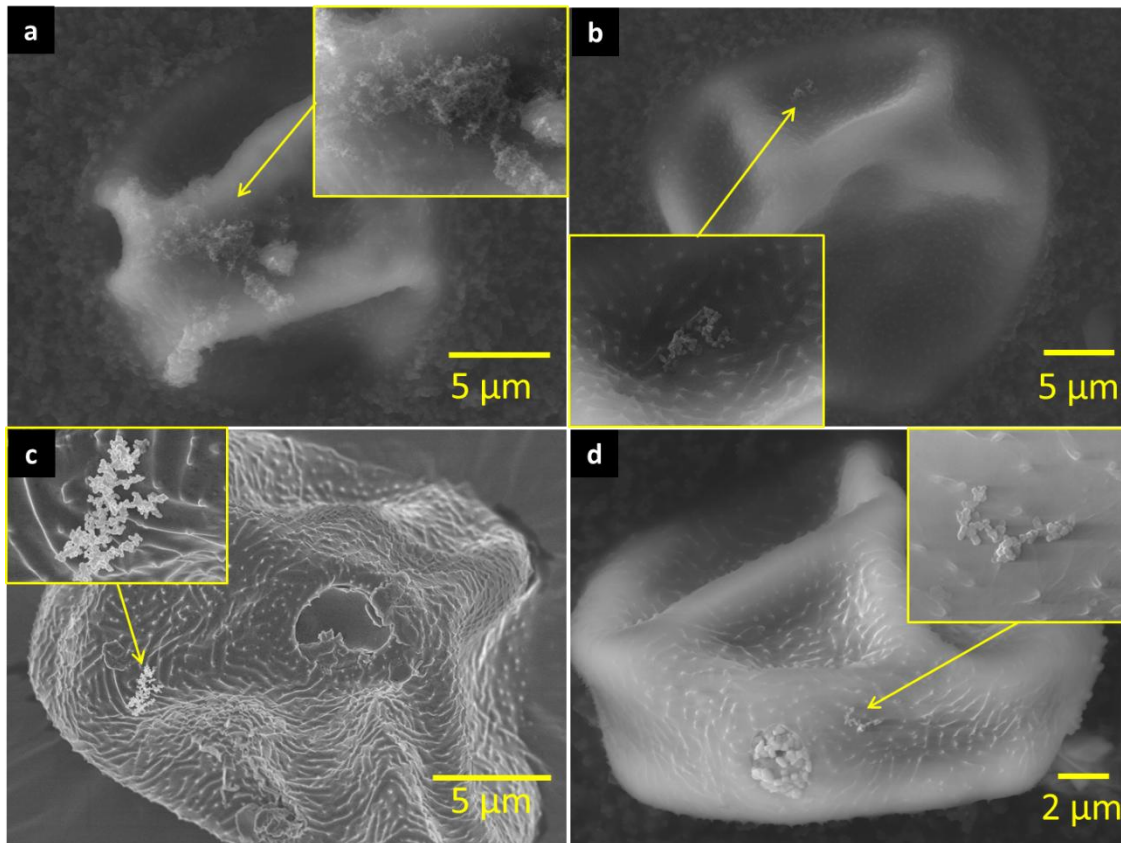


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181 Figure 3. SEM images of polluted birch pollen grains carrying: (a) 1 μm-sized calcium silicate particle;  
182 (b) aluminosilicate particles; (c) a large soil dust particle (plagioclase Al-Si-Ca-Fe-rich); (d) a large  
183 phyllosilicate particle,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

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

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



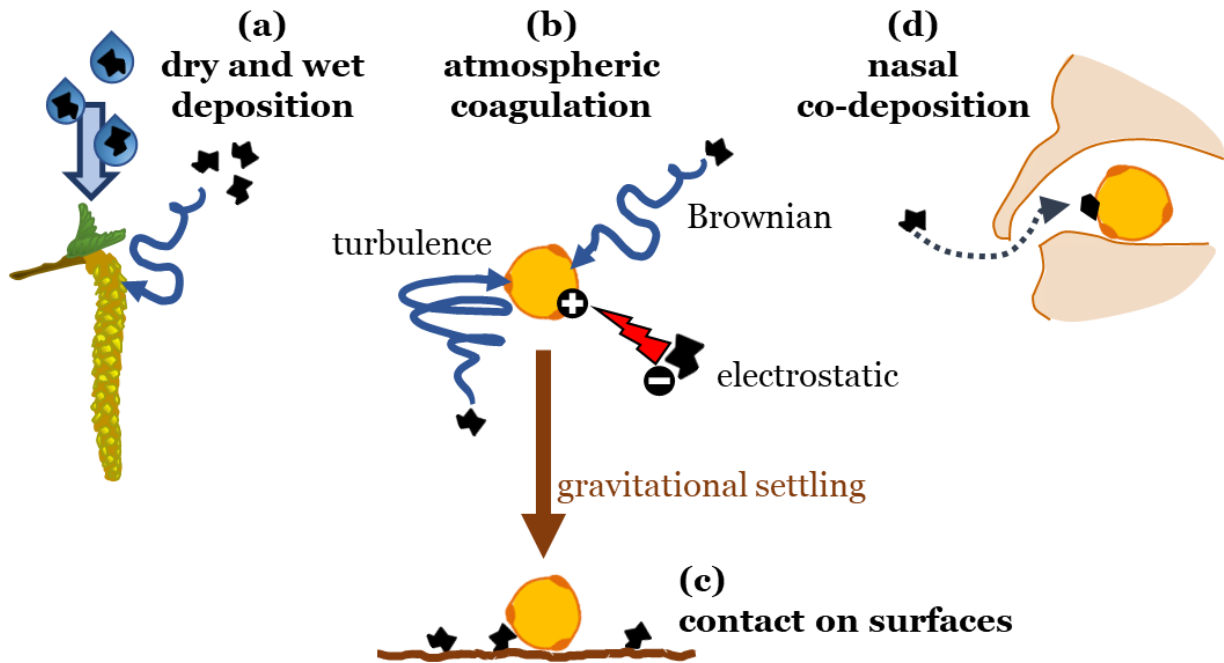
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191 Figure 4. SEM images of polluted birch pollen grains: (a) covered with a large chain-like soot aggregate  
192 of nanospheres (diameter 40-50 nm); (b-c) carrying a micrometer-sized chain-like soot aggregate of  
193 nanospheres (diameter 80-90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

#### 195 **4.DISCUSSION**

196 Our atmospheric samples collected under conditions of moderate particulate air  
197 pollution reveals that pollution of the surface of pollen grains (PGs) by atmospheric  
198 particles is a common phenomenon: out of a total of 236 birch pollen grains (BPGs)  
199 examined, 32% of BPGs carried an average of  $2 \pm 1$  particles (205 particles deposited  
200 on 75 BPGs). On a global average during our sampling, BPGs carried 0.86 particles  
201 (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 PM<sub>10</sub> during sampling (Choël  
213 et al., 2020). The use of a PM<sub>10</sub> 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 (PM<sub>10</sub> = 26.3 ± 8.3 µg·m<sup>-3</sup>), 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.



226

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

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

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

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

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

Equation 1		N particle concentration (#.m <sup>-3</sup> )
<b>Rate of collision</b>	$J_{12} = K_{12}N_1N_2$	$K_{12}$ coagulation rate coefficients
Equation 2		k Boltzmann constant; $\mu$ viscosity of air (20°C, 1.825×10 <sup>-5</sup> kg·m <sup>-1</sup> ·s <sup>-1</sup> ); T temperature (K); $D_p$ diameter of particles (m); $C_c$ Cunningham correction factor
<b>Brownian coagulation</b>	$K_{12}^{BG} = \frac{2kT}{3\mu} (D_{P1} + D_{P2}) \left( \frac{C_{C1}}{D_{P1}} + \frac{C_{C2}}{D_{P2}} \right)$	
Equation 3		$\vartheta_t$ terminal settling velocity (BPGs = 0.012 m·s <sup>-1</sup> ((Sofiev et al., 2006))); $E(D_{P1}, D_{P2})$ collision efficiency (=1 in this work)
<b>Gravitational coagulation</b>	$K_{12}^{GC} = \frac{\pi}{4} (D_{P1} + D_{P2})^2 (\vartheta_{t1} - \vartheta_{t2}) E(D_{P1}, D_{P2})$	
Equation 4		$\varepsilon_k$ rate of dissipation of kinetic energy per unit mass (10 s <sup>-1</sup> ); $\nu$ kinematic viscosity of fluid = (15.06×10 <sup>-6</sup> m <sup>2</sup> ·s)
<b>Coagulation in turbulent flow</b>	$K_{12}^{TF} = \left( \frac{\pi^2 \varepsilon_k}{120\nu} \right)^{1/2} (D_{P1} + D_{P2})^3$	

251

252 Calculations of  $K_{12}$  for the three types of coagulation described in Table were  
 253 performed for a particle deposition on BPG (21  $\mu$ m in diameter) from a monomodal  
 254 population of 2  $\mu$ m diameter particles at a concentration of 5 particules·cm<sup>-3</sup>,

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

267 Under typical pollination weather conditions (absence of turbulence, calm weather  
268 and low wind), and without taking into account electrostatic interactions between  
269 particles, the main coagulation process would be the deposition of particles by  
270 gravitational settling. By assuming a concentration of  $2\text{-}\mu\text{m}$ -sized particles  $N_2$  in the  
271 order of  $5 \text{ particles}\cdot\text{cm}^{-3}$  and a gravitational settling time of one hour (drop of  $43 \text{ m}$ ),  
272 we calculate  $0.09$  particles deposited per PG ( $J_{12} = 2.4 \times 10^{-5} \text{ particles}\cdot\text{s}^{-1}$ ). For pollen  
273 transported over long distances, while some PGs have been observed at considerable  
274 altitudes of  $2 \text{ km}$  (Damialis et al., 2017), it is conceivable that gravitational settling  
275 alone explains the levels of particulate contamination of PGs observed in our samples  
276 (on average  $0.86$  particles per BPG). If the hypothesis of coagulation primarily by  
277 gravitational settling is correct, then the presence of particles on the surface of BPGs  
278 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  $\mu\text{m}$  range, with a  
283 maximum around 50 to 150  $\mu\text{m}$  (Xie et al., 2009). Our estimates show that the  
284 encounter between a PG and a coarse particle of about 100  $\mu\text{m}$  will be very unlikely  
285 for contact times of a few minutes. For fine particles (2  $\mu\text{m}$ ) exhaled at a  
286 concentration of 150,000 particles $\cdot\text{m}^{-3}$  in contact with a high concentration of BPGs  
287 (1,000 BPGs $\cdot\text{m}^{-3}$ ), the calculated deposition rate of 2- $\mu\text{m}$ -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  $\text{cm}^3$  (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

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

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

## 347 **5.CONCLUSION**

348 An average pollen surface contamination of  $2 \pm 1$  particles per BPG was obtained for  
349 38% of BPGs sampled under moderate air pollution conditions (average  $PM_{10} = 26.3$   
350  $\mu\text{g}\cdot\text{m}^{-3}$ ). This value was not known in the literature because previous estimates were  
351 possibly subject to sampling artifacts. Determination of this particulate  
352 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  $\mu\text{m}$  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  $\mu\text{m}^2$ , 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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