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1	Quantitative Assessment of Coagulation of Atmospheric		
2	Particles onto Airborne Birch Pollen Grains		
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20 ABSTRACT

The coagulation of airborne particles on the surface of allergenic pollen grains is 21 poorly described in the literature. However, particles deposited on the surface of 22 pollen grains could have an effect on allergy and its symptoms. Observations of pollen 23 surface alterations reported in the literature are either qualitative or are tainted by 24 sampling artifacts that overestimate surface particulate pollution. Birch pollen grains 25 (BPGs) were sampled in the atmosphere during pollination to quantitatively assess 26 the extent of particulate deposition on allergenic pollen surfaces. Airborne PGs were 27 28 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 29 particles adhering on pollen surfaces were performed by SEM/EDX. 68% of PGs did 30 not carry any particle on their visible surface while the remaining 32% had between 1 31 to 16 particles on their surface. On average, polluted BPGs carried 2 ± 1 particles, 32 representing an average surface coverage of 0.95%. We hypothesized that the main 33 coagulation process was particle deposition by gravitational settling. Collision 34 velocity calculations indicate that coagulation due to gravitational settling is 35 prevalent under typical pollination atmospheric conditions. However, the effects of 36 electric charges carried by pollen are largely unknown and may significantly alter 37 coagulation rates. The presence of particle-polluted allergenic pollens appears to be 38 common in the urban atmosphere, even under low air pollution conditions. The 39 impact of chemical pollution of BPGs due to the presence of particulate matter 40 (including soot particles) remains relatively unknown, both on the allergy and on the 41 42 reproductive capacity of the tree.

43

45 1.INTRODUCTION

Birch pollen grains (BPGs) cause allergic reactions in more than 100 million people 46 worldwide (Lavaud et al., 2013). An increasing trend in atmospheric BPG 47 concentrations has been observed in several countries in relation to the relentless rise 48 of carbon dioxide and the effects of climate change (Besancenot et al., 2019; Darbah 49 et al., 2008; Frei, 1998; Hoebeke et al., 2017; Ziska et al., 2019). In addition, air 50 pollution can aggravate allergy symptoms (Robichaud, 2021; Schiavoni et al., 2017). 51 The allergic patient indeed never breathes BPGs alone; airborne BPGs are suspended 52 in the urban atmosphere consisting of gaseous pollutants (NO₂, O₃, Volatile Organic 53 Compounds), coarse, fine and ultrafine particles and other bioaerosols (spores and 54 other potentially allergenic pollens). Taken separately, each of the constituents of the 55 urban atmosphere can have aggravating effects on allergy but physical-chemical 56 interactions also exist between gaseous and particulate atmospheric compounds. 57 Alterations of BPGs due to atmospheric biochemical reactions with ozone or nitrogen 58 dioxide have been highlighted (Sénéchal et al., 2015). Gaseous air pollutants modify 59 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 clear qualitative evidences that allergenic PGs can also act as carriers of pollutants, 62 63 both adsorbed gases and coagulated particles (Okuyama et al., 2007; Zhu et al., 64 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 70 the pollen surface of 16 \pm 12 (average \pm standard deviation), 39 \pm 32 and 4 \pm 4 71 particles per PG for Acer, Pinus and Platanus pollen, respectively (Ribeiro et al., 72 2015). This study was conducted using a Hirst volumetric spore trap which is the 73 most common method of sampling pollen on a moving slide, originally developed to 74 count ambient pollen (Mullins & Emberlin, 1997). However, the use of a Hirst-type 75 pollen sampler induces a sampling artifact with a significant overestimation of 76 particle adhesion on PGs (Choël et al., 2020). In addition, to our knowledge, there are 77 no quantitative data on the specific pollution of the surface of BPGs by atmospheric 78 particulate matter (APM); only one study reported a high degree of particle 79 agglomeration onto BPGs collected from two highly polluted urban areas (Behrendt 80 81 et al., 1991).

Being able to sample windborne PGs while preserving their initial state is crucial to 82 study how PM alters the physical, chemical and biological properties of PGs. PM 83 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 efficiently trapped in a specific sampling stage (for coarse particles) different from 86 87 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, 88 compared to active sampling pollen trap, such as Hirst-type samplers (Choël et al., 89 2020; Ribeiro et al., 2015; Wittmaack et al., 2005). In addition, the sampling 90 duration should be short enough (~a few hours) to avoid the pile-up of particles, 91 especially in highly polluted environments (Wittmaack et al., 2005). The objective of 92 this work is to document and reliably assess the size range, particle types and 93 frequency of occurrence of particles adhered to airborne BPGs sampled under fairly 94

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

Pollen sampling was carried out daily from 30 March to 7 April 2017 (start and end 100 time: 10:00 am) on the roof of a building of Lille University campus (Lat. 50° 36' 29" 101 N, Long. 3° 8' 25" E, Altitude 20 m a.s.l.) (Supplementary Figure So) when weekly 102 cumulative birch pollen concentrations peaked at 3022 grains-m⁻³ according to the 103 French monitoring network (RNSA) in Lille metropolitan area (Figure 1). On 30 and 104 31 March 2017, daily birch pollen concentrations peaked at 986 and 1236 BPGs per 105 m³, respectively. Meteorological parameters were recorded by a weather station 106 (Davis Vantage Pro) installed at the sampling site (Supplementary Figure S1). 107



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 µg·m⁻³) and in red dotted line for PM2.5 (25 µg·m⁻³).

No rain event occurred during the study period. Birch dehiscence was triggered by a 113 temperature rise to 16°C combined with a drop of relative humidity on 30 March. The 114 weekly average temperature, relative humidity, and wind speed were 11.9 \pm 2.2 °C, 115 76.7 \pm 5.1 % RH, 1.4 \pm 0.5 m·s⁻¹, respectively. Ambient PM₁₀ and PM_{2.5} concentrations 116 were 26.3 \pm 8.3 µg·m⁻³ and 19.3 \pm 8.2 µg·m⁻³, respectively at the closest air quality 117 monitoring station located 4 km away from the pollen sampling site (public data 118 provided by ATMO Hauts-de-France). During the study period, PM₁₀ concentrations 119 never exceeded the WHO recommended threshold value, i.e. 50 µg·m⁻³ (24-hour 120 mean). In contrast, PM_{2.5} concentrations were over the WHO guideline of 25 µg·m⁻³ 121 (24-hour mean) on 3 and 4 April 2017 (Figure 1). Airborne PGs were trapped on the 122 first stage PM>10 µm of a 3-stage cascade impactor (PM10 impactor, Dekati Ltd.), i.e. 123 impaction stage with particle size cut-off diameter of 10 µm at 50% collection 124 efficiency valid for spherical particles with a mass density of 1 g·cm⁻³. The impactor 125 was mounted on the top of a 4-meter-high mast (Supplementary Figure S2). Airborne 126 PGs were impacted directly on a conductive carbon tape for electron microscopic 127 examination. Bounce-off and re-entrainment of deposited PGs from the impaction 128 surfaces can be minimized by using adhesive collection substrates (Wittmaack et al., 129 2005). 130

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).



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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 150 substrate appear with similar grey levels. To confirm the presence of a BPG and to 151 examine particulate adhesion, each BPG was magnified and secondary electron (SE) 152 images sensitive to topography were taken. Counting and sizing of particles adhering 153 to pollen surfaces were determined from BSE images using FIJI/ImageJ software. X-154 ray elemental analysis of particles attached to PGs was carried out with an energy-155 dispersive X-ray (EDX) detector (SDD 80 mm², Oxford Instruments Inc.) at an 156 accelerating voltage of 15 kV. Note that BPGs appear deflated as a result of shrinkage 157 due to low-vacuum which enables EDX analysis of micrometer-sized particles down 158 to 0.2 µm (Choël et al., 2010). 159

160 **3.RESULTS**

161 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 162 particle on their visible surface. The remaining 32% of PGs had between 1 and 16 163 particles adhered to their surface. On average, polluted PGs had 2 ± 1 particles on 164 their surface. Sampling artifact was assessed to 2.2 ± 2.4 particles per PG with a 165 166 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) 167 particles per BPG) confirms the need to use an impactor because the sampling 168 artifact of an HTS is of the same order of magnitude. 169

Particles deposited on BPGs had an average diameter of $1.5 \pm 2.0 \ \mu m$ (average \pm 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 $5.6 \pm 4.0 \ \mu m$ 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).



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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).

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



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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.

195 **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

covered by particles is however very low, less than 1%, which contrasts with the 202 higher fractions previously reported in the literature (Behrendt et al., 1991; Ribeiro et 203 204 al., 2015). For example, the area covered by particles on pine and maple pollen collected from the atmosphere with a Hirst trap was 5.7% and 7.5%, respectively 205 (Ribeiro et al., 2015). A very high degree of agglomeration was also observed for BPG 206 collected with a high volume sampler in highly polluted urban areas (Behrendt et al., 207 1991). We assume that the high degrees of agglomeration presented in these previous 208 works may be subject to a sampling artefact involving an overestimation of the 209 number of particles per PG. Hirst-type samplers are not suitable for the assessment 210 211 of particulate pollution on the surface of PGs due to a too high impaction velocity which does not allow the discrimination of pollen and PM10 during sampling (Choël 212 et al., 2020). The use of a PM10 impactor avoids this sampling artefact and explains, 213 214 at least in part, the lower particulate pollution rate obtained in this study (Choël et al., 2020). Another explanation is that the air quality was relatively good in our study 215 $(PM10 = 26.3 \pm 8.3 \mu g \cdot m^{-3})$, which reduces the probability of coagulation of 216 atmospheric particles on the surface of PGs. One of the limitations of our study is the 217 218 low number of PGs observed. Manual grain-by-grain observation by electron microscopy is indeed time-consuming and unsuitable for samples of several thousand 219 grains but it is the only way to reliably assess the presence of particles on PGs. In 220 addition, some particles are difficult to identify, in particular due to the presence of 221 sub-pollen particles on the surface of BPGs (Ruggiero & Bedini, 2018, 2020). It 222 should also be remembered that SEM analysis only allows to observe the visible 223 pollen surface; particles stuck to the pollen surface on the side of the collection 224 225 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	$J_{12} = K_{12} N_1 N_2$	N particle concentration (#.m ⁻³) K ₁₂ coagulation rate coefficients
Equation 2 Brownian coagulation	$K_{12}^{BG} = \frac{2kT}{3\mu} (D_{P1} + D_{P2}) \left(\frac{C_{C1}}{D_{P1}} + \frac{C_{C2}}{D_{P2}}\right)$	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	$K_{12}^{TF} = \left(\frac{\pi^2 \varepsilon_k}{120\upsilon}\right)^{1/2} (D_{P1} + D_{P2})^3$	\mathcal{E}_k rate of dissipation of kinetic energy per unit mass (10 s ⁻¹); υ kinematic viscosity of fluid = (15.06×10 ⁻⁶ m ² ·s)

252 Calculations of K_{12} for the three types of coagulation described in Table were 253 performed for a particle deposition on BPG (21 µm in diameter) from a monomodal 254 population of 2 µm diameter particles at a concentration of 5 particules cm⁻³,

considering a terminal settling velocity of 100 cm \cdot h⁻¹ for particles (1.2 cm \cdot s⁻¹ for BPG) 255and no rebounds after collision. This computational estimate shows that K_{12} is of the 256 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 257 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 258 negligible ($K_{12}^{BG} \approx 2 \times 10^{-15} \text{ m}^6 \cdot \text{s}^{-1}$). Equation 3 tends to predict very high coagulation 259 with the finer particles, as the difference in settling velocity is greater and the 260 261 concentrations of the fine particles are several thousands to tens of thousands of particles per cm³. However, the efficiency of collision E_{12} , which depends on the size 262 of the particles involved, decreases sharply as the diameter of particle 2 tends 263 towards zero (Seinfeld & Pandis, 2016). This prediction is consistent with our 264 observations where relatively few very fine particles (< 500 nm) were detected on the 265 266 surface of BPGs.

267 Under typical pollination weather conditions (absence of turbulence, calm weather and low wind), and without taking into account electrostatic interactions between 268 particles, the main coagulation process would be the deposition of particles by 269 gravitational settling. By assuming a concentration of 2- μ m-sized particles N_2 in the 270 order of 5 particles.cm⁻³ and a gravitational settling time of one hour (drop of 43 m), 271 we calculate 0.09 particles deposited per PG ($J_{12}=2.4\times10^{-5}$ particles·s⁻¹). For pollen 272 transported over long distances, while some PGs have been observed at considerable 273 altitudes of 2 km (Damialis et al., 2017), it is conceivable that gravitational settling 274 alone explains the levels of particulate contamination of PGs observed in our samples 275 276 (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 277 could be a marker of atmospheric transport of pollen over long distances. 278

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

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

coagulation because, on the one hand, airborne particle concentrations can be as high 304 as 10⁶ to 10⁷ particles per cm³ (Seinfeld & Pandis, 2016) and, on the other hand, 305 pollen concentrations can be higher (Charalampopoulos et al., 2021). Further 306 sampling, especially at street level, is therefore necessary to better assess the fraction 307 of PGs polluted by soot, especially as a large body of literature highlights the 308 309 aggravating effects on allergy of co-exposure to pollen and soot (Chen et al., 2020; Nel et al., 1998; Shiraiwa et al., 2012). Unlike nasal co-deposition (Figure 5), the 310 pollen and soot (or pollen and particles) mixture resulting from atmospheric 311 coagulation may have time (up to several days) to undergo atmospheric aging. 312 Chemical reactivity at the particle-pollen contact surface cannot be excluded. Very 313 little information is available on this multiphase pollen chemistry; for example, it has 314 been shown that in the presence of moisture, allergens can diffuse from the PG to 315 316 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 will be whether the level of particle deposition observed in this study on airborne 320 BPGs (0.86 particles per PG) is significant in comparison with the effects of co-321 exposure to pollen and APM during breathing (Figure 5d). First, it should be noted 322 that the effects of these two mechanisms are cumulative; co-deposition also occurs on 323 PGs that have already been subjected to coagulation of atmospheric particles. In 324 addition, and as previously mentioned, atmospheric aging involving at least 325 biochemical reactivity and diffusion of allergens may occur after coagulation during 326 atmospheric transport of BPGs. Our study alone cannot determine the potential 327 impacts of pollen surface particle pollution on allergy. However, our atmospheric 328

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%).

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

347 5.CONCLUSION

An average pollen surface contamination of 2 ± 1 particles per BPG was obtained for 349 38% of BPGs sampled under moderate air pollution conditions (average PM10 = 26.3 350 µg·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 provides an estimate of a realistic dose-response effect of APM coagulated on pollensurface.

BPGs are small (21 µm in diameter) and larger pollens could have higher APM 355 356 deposition rates. Gravitational settling is indeed suspected be the predominant mechanism of coagulation in the atmosphere. Very little is known about pollen 357 pollution by APM on the plant, about electrostatic interactions between particles and 358 also about pollen resuspension after deposition on a surface. APM coagulation on 359 360 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.

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

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