



Evaluating inaccurate pollen concentrations caused by turbulence using passive sampler

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Received: 16 April 2021 / Accepted: 28 September 2021
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Abstract Understanding the atmospheric concentration of pollen (which is one of the most common vectors of allergens) is necessary to determine the atmospheric allergen level. Airborne pollen concentrations were predominantly evaluated using volumetric and gravitational particle samplers. However, no previous studies have successfully shown whether these sampling methods can be used to evaluate actual airborne pollen concentrations. In this study, the relationship between wind speed and sampling efficiency was investigated to determine whether the influence of ambient wind speed on sampling efficiency was significant. To this end, the influence of wind speed was analyzed by comparing a volumetric sampler and a gravitational sampler. The experimental

results indicate that an increase in the wind speed results in an increase in the sampling efficiency of the gravitational sampler because of fluctuations in the turbulence. Our simulation shows that when pollen is entrained in the turbulence, the changes in the wind speed, turbulence amplitude, and turbulence length influence the deposition rate of pollen on the gravitational sampler. These influences can be explained by the turbulence vibration model. These results show the inadequacy of existing evaluation methods, not only for pollen deposition data, but also for all types of bioaerosol deposition data.

Keywords Pollen sampling · Volumetric sampler · Gravitational sampler · Actual airborne concentration · Turbulence

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1 Introduction

Since pollen distribution information is important to avoid unnecessary exposure to pollen, various pollen sampling methods have been proposed to determine airborne pollen concentrations. However, the sampling methods have different sampling efficiencies. Thus, various researchers have been working on the relationship between sampling efficiency and sampling methods (e.g., Bhat & Rajasab, 1989; Crisp et al., 2013; Fiorina et al., 1997; Heffer et al., 2005;

Jetschni & Jochner-Oette, 2021; Miki et al., 2017, 2019; Tomás et al., 1997).

Among the various sampling devices proposed, comparison of volumetric samplers and gravitational samplers is very important for the quality control of allergen monitoring. In order to understand the health impact of allergic pollen on humans, volumetric samplers, which are designed to imitate respiratory function, have been widely used for pollen sampling (e.g., Camacho et al., 2017; Carinanos et al., 2002; Hirst, 1952; Hrga et al., 2010; Monroy-Colin et al., 2018; Moreno-Grau et al., 1998; Perez-Badia et al., 2013; Peternel et al., 2003; Ribeiro et al., 2008). The gravitational method is another method used frequently; it is employed not only to understand the deposition quantity of pollen but also because of its simplicity (e.g., Durham, 1946; Hofmann et al., 2016; Tosunoğlu et al., 2013; Tseng et al., 2019). Gravitational samplers are also used to understand pollen deposition on the skin of patients (Erkara et al., 2009).

The relationship between the sampling efficiencies of gravitational and volumetric methods has been studied to determine the amount of inhaled allergens from pollen deposition information (e.g., Hassan & Lau, 2009; Ranta et al., 2008).

Previous studies have shown that pollen concentrations evaluated by volumetric samplers and passive samplers have clear correlations (Bricchi et al., 2000; Sun et al., 2017). Conversely, it has been stated that volumetric and gravitational methods have virtually no correlation (Cornell et al., 1961).

In this study, the two most frequently used pollen sampling methods—a volumetric method called the Hirst sampler, which is the most commonly used volumetric sampler, and a gravitational method called the Durham sampler, which is the most commonly used gravitational sampler, were compared to reveal the conditions that make the relationship between the two methods unsteady.

2 Materials and methods

2.1 Experimental set-up

The Hirst sampler and Durham sampler (Fig. 1) were permanently set on the rooftop of the Hokkaido Institute of Public Health (43° 4'59'' N, 141° 19' 59'' E). The sampling rate of the Hirst sampler

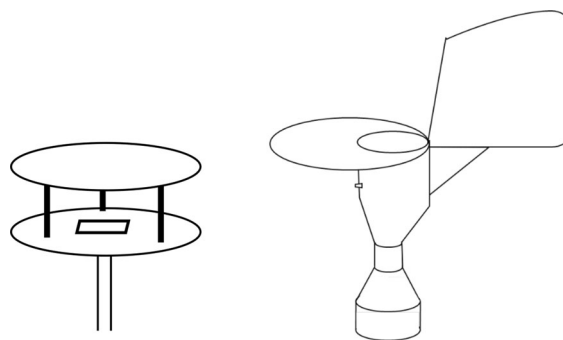


Fig. 1 Schematics of the Hirst sampler (right) and Durham sampler (left)

(Burkard Manufacturing Co. Ltd., Rickmansworth, UK) was set to 10 L min^{-1} . Pollen grains sampled by these devices were counted and sorted through visual observation using a microscope (Olympus BX 53-F, Tokyo, Japan). Sampling by the Hirst sampler was performed following the IAA protocol (Galán et al., 2014), and the Durham sampler was performed following the standardized protocol (PAAA and IAA protocol).

Although all pollen taxa in the atmosphere were sampled, *Betula* was chosen as the study object because of its representativeness as the main allergenic pollen. Considering the main *Betula* pollen season (Inatsu et al., 2014), *Betula* pollen sampling was performed from April 20–May 20 annually, from 2013 to 2016, and the daily pollen concentration values were analyzed. Since forests were situated to the north of the sampling site, in order to obtain clear data without background errors, we used data collected, only when the daily dominant wind direction was not from the north. Additionally, some daily data were missing because of technical problems with the Hirst sampler.

2.2 Theory

A method to determine the airborne pollen concentration from sampling results was first introduced by Scheppegrell (1917) and subsequently improved by Cocke (1937) and Feinberg and Steinberg (1933) as follows:

$$N = v_s \times a \times t \times C_G, \quad (1)$$

where N is the number of deposited pollen grains, v_s is the pollen settling speed (m s^{-1}), a is the sampling

area (m^2), t is the sampling duration (s), and C_G is the atmospheric pollen concentration (grains m^{-3}). For v_s , the terminal velocity derived from Stokes' law is usually substituted, which is calculated as:

$$v_s = \frac{2r_p^2(\rho_p - \rho_f)g}{9\eta}, \quad (2)$$

where r_p is the radius of a particle, ρ_p is the density of the particle, ρ_f is the density of the fluid, and η is the viscosity of the ambient fluid. Because the radius of a pollen grain varies, a representative pollen grain radius that fits the experiment best was calculated and substituted; specifically, $r_p = 17(\mu\text{m})$. Under the assumption that laminar flow is dominant in the sampling site, the particle settling speed is only dependent on the particle size (r_p) because streamlines do not intersect with each other in the laminar flow.

We used these equations to convert the gravitational sampling results from the Durham sampler to the airborne pollen concentration. For the density of pollen grains, 1 g cm^{-3} was substituted. After the conversion, the relationship between the sampling efficiency of the gravitational sampler and that of the volumetric sampler was analyzed using linear regression and Pearson correlation coefficients.

3 Results and discussion

The airborne pollen concentration data from the gravitational sampler and the volumetric sampler exhibited high correlations, especially in 2013 and 2016, showing that in these two years, the identification and counting of *Betula* pollen for both the gravitational sampler and the volumetric sampler were performed precisely, whereas the correlation coefficients for 2014 and 2015 were relatively low ($r = 0.999$ (2013); $r = 0.767$ (2014); $r = 0.850$ (2015); $r = 0.986$ (2016)) (Fig. 2). Only a limited amount of data are available for 2013 because volumetric sampling was performed only from May 9 because of a technical problem. The slopes of the scatter plots of the volumetric sampling data and the converted gravitational sampling data show differences in sampling efficiencies, which are ratios of the sampling efficiency of the gravitational sampling efficiency to that of the volumetric sampling efficiency. These results indicate that the converted

gravitational sampling results tend to exhibit higher airborne pollen concentrations than the volumetric sampling method.

The correlations between the sampling ratio of the gravitational method to the volumetric method and the ratio of wind speed, which is a significant meteorological factor for particle sampling, to the sampling rate, which was constant, are also shown in scatter plots (Fig. 3). In these plots, proportional relations are shown ($r = 0.745$ [2013]; $r = 0.696$ [2014]; $r = 0.407$ [2015]; $r = 0.627$ [2016]).

The results show that the ratio of the sampling efficiency of the gravitational sampler to that of the volumetric sampler is proportional to an increase in the wind speed, which means that the conventional conversion method cannot be used to calculate the volumetric pollen concentration from pollen deposition data on account of the influence of wind speed. These results show that in some cases, inaccurate conversion between the gravitational sampler and the volumetric sampler is performed either because the volumetric sampling efficiency decreases with an increase in wind speed or because the gravitational sampling efficiency increases with an increase in the wind speed.

3.1 Inadequacy of existing correction theory for volumetric sampler

The sampling efficiency is inherently independent of the wind speed, which is widely assumed in the discussion of sampling efficiency. This is derived from the following equations:

From the continuity equation for particles,

$$C_0Au = C_Sau_s, \quad (3)$$

where C_0 is the actual airborne pollen concentration, A is the cross-sectional area of the limiting trajectory, u is the wind speed, C_S is the evaluated airborne pollen concentration, a is the cross-sectional area of the sampling tube, and u_s is the sampling rate. In addition, the continuity equation of air is

$$Au = au_s. \quad (4)$$

Thus, from Eq. (3) and Eq. (4), the following result is derived, which is assumed to be true for any type of airborne particle sampling:

$$C_0 = C_S. \quad (5)$$

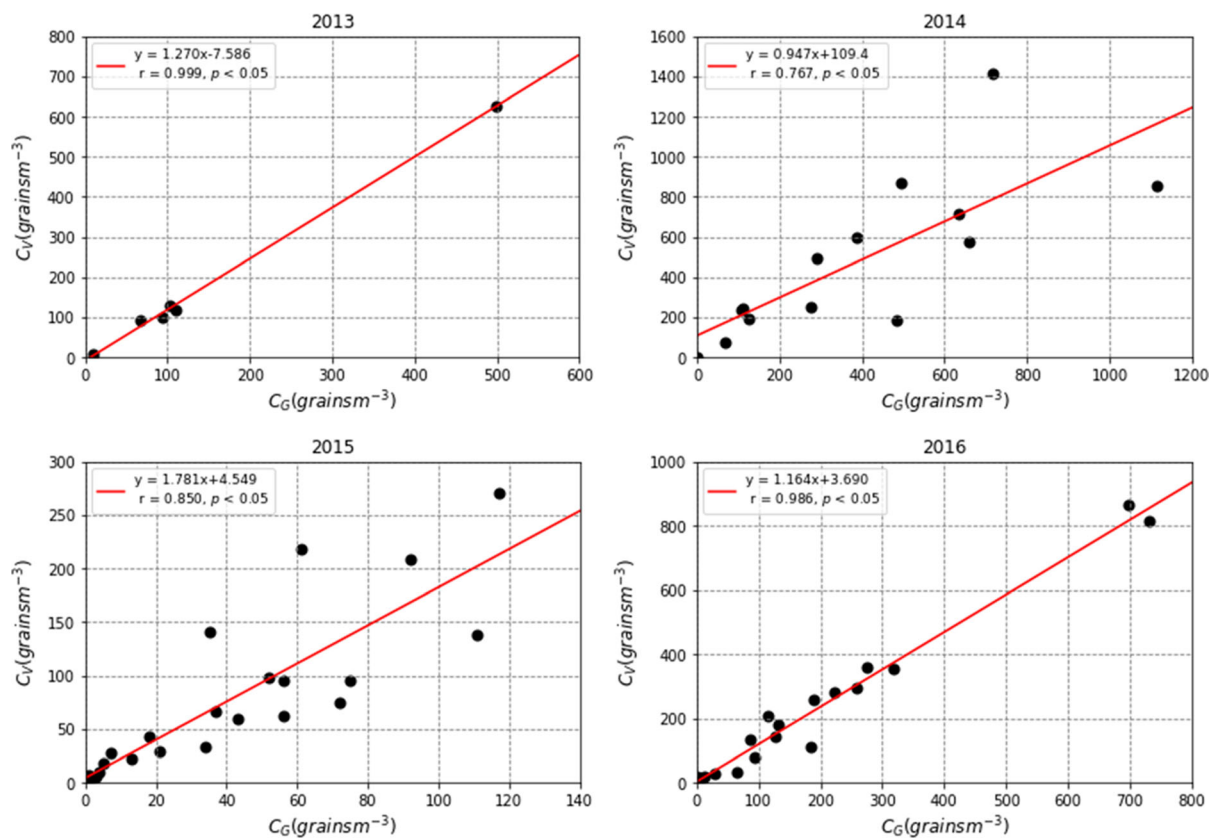


Fig. 2 Relationship between airborne pollen concentration evaluated from volumetric (C_V) and gravitational (C_G) sampling results

However, previous studies have suggested that the relationship between the sampled airborne particle concentration and the actual airborne particle concentration is a function of wind speed. Thus, in order to provide for the influence of wind speed on the sampling efficiency of volumetric samplers, a correction theory was introduced by Davies (1968) and improved by Vincent (2007). According to Davies theory, the changes in the relative wind speed of ambient air to the sampling rate proportionally influence the changes in the sampling efficiency because of changes in the inertial force that affect the particles, which is calculated as follows:

$$\alpha = 1 + \beta(R - 1), \quad (6)$$

$$\beta = 1 - \left\{ \frac{1}{1 - 0.6St \ln(1 - \frac{\delta}{2})} \right\}, \quad (7)$$

where α is the sampling efficiency, $R = u_s/u$, St is the Stokes number, and δ is the diameter of the sampling

tube. These equations (Eqs. (6) and (7)) demonstrate the possibility that a particle inside the limiting trajectory could be pushed out of the limiting trajectory when the ambient wind speed was faster than the sampling rate. Meanwhile, for sampling rates faster than the ambient wind speed, there is a chance that a particle outside the limiting trajectory can be pushed over the limiting trajectory. α in Eq. (6), representing the theoretical probability of the occurrence of these crossovers. The Davies theory has been significantly enhanced and applied in various studies. However, the results from the present study show a reverse trend, in which an increase in the wind speed causes an increase in the evaluated airborne pollen concentration. Thus, it is reasonable to say that additional influential factors resulted in a linear relationship between wind speed and the ratio of the sampling efficiency of the gravitational method when compared to that of the volumetric method. We assumed that the additional influential factor was related to the interpretation of the gravitational sampling results.

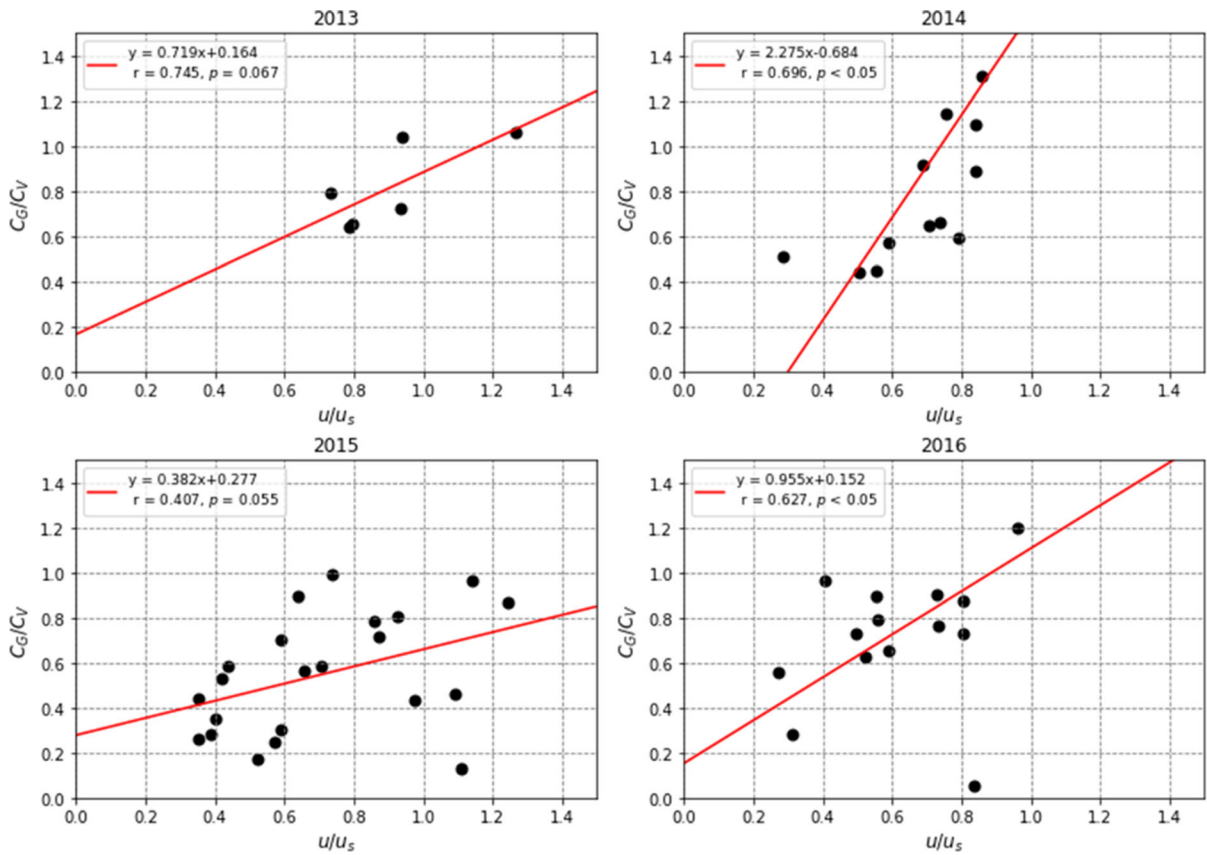


Fig. 3 Relationship between the ratio of evaluated airborne pollen concentration from gravitational sampling (C_G) to that from volumetric sampling (C_V) and the ratio wind speed (u) to sampling rate (u_s)

3.2 Consideration of the influence of turbulence on gravitational sampling method

Although the particle deposition speed was assumed to be the same as the settling speed above because the simple conversion has been often used, more sophisticated particle deposition theories also have been established. These theories were summarized by Seinfeld and Pandis (2016) as follows:

$$v_d = \frac{1}{r_a + r_b + r_a r_b v_s} + v_s, \quad (8)$$

$$r_a = \frac{1}{\kappa u_*} \left[\ln \left(\frac{z}{z_0} \right) \right], \quad (9)$$

$$r_b = \frac{1}{3u_* \left[Sc^{-\gamma} + \left(\frac{St}{C_1 + St} \right)^2 + \frac{1}{2} \left(\frac{2r_p}{C_2} \right)^2 \right] R_1}, \quad (10)$$

$$R_1 = \exp(-St^{\frac{1}{2}}), \quad (11)$$

where v_d is the particle deposition speed, u_* is the friction velocity, r_a is the aerodynamic resistance, r_b is the quasi-laminar layer resistance, γ is the value depending on the roughness of the surface, z_0 is the roughness length, z is the reference height, Sc is the Schmidt number, C_1 and C_2 are the parameters depending on the land use, κ is the von Karman constant, and R_1 is the correction factor.

Under the assumption that the temperature profile above the sampling plate is neutral, the settling speed of a particle increases with an increase in wind speed. However, since the conventional particle deposition theory for gravitational samplers was established under the assumption that laminar flow is dominant, the conventional method may no longer be applicable when the turbulence is the dominant airflow at the sampling site.

The dominant flow state, laminar flow, or turbulent flow at the sampling site was determined by the Reynolds number. The Reynolds number for the sampling condition in this experiment is calculated by substituting 4.2 m s^{-1} (which is the average wind speed for the duration of the experiment) for the wind speed and $3.0 \times 10^{-1} \text{ m}$ (which is the diameter of the laminas of the Durham sampler (d)) for the characteristic length in Eq. (12).

$$Re = \frac{\rho u d}{\eta} = \frac{1.25 \times 4.2 \times 3.0 \times 10^{-1}}{1.5 \times 10^{-5}} \approx 1.05 \times 10^5. \quad (12)$$

Since the Reynolds number is much larger than 4.0×10^3 , the dominant flow at the sampling site was turbulent for the duration of sampling. Thus, it is reasonable to say that since the theoretical airborne pollen concentration was calculated from the pollen deposition data using Eqs. (1) and (2) while assuming laminar flow, discrepancies were present between the theoretically calculated airborne pollen concentrations and the actual pollen concentrations at the sampling site where turbulent flow was dominant. Although pollen dispersion by turbulence has been discussed in previous studies (Chamecki & Meneveau, 2011; Sabban & van Hout, 2011; Timerman et al., 2014), the direct influence of turbulence on the gravitational sampling efficiency is rarely known.

As mentioned above, the results show that the existing calculation method is not applicable to turbulent environments, although the conversion method of pollen deposition data to airborne pollen concentration in turbulent flow has rarely been considered. The most characteristic difference between laminar and turbulent flows is that the flow lines in turbulent flow vibrate, whereas the flow lines in laminar flow maintain a steady state.

3.2.1 Building structure of turbulence simulation

A preliminary two-dimensional simulation was performed under the following conditions in order to evaluate the influence of turbulence on pollen deposition. A sampling plate with a length of 1 m was placed at the coordinate origin. Pollen grains with a radius of $10 \mu\text{m}$ and density of 1000 kg m^{-3} were

randomly scattered with an airborne concentration of 10 grains m^{-3} , and were flown from the positive to the negative direction of the horizontal axis (Fig. 4). The size of the pollen grains in the simulation was set to $10 \mu\text{m}$ because of the representativeness of the pollen size. Pollen particles are driven by friction from wind, buoyancy, and gravity. Through fluctuations caused by turbulence, every pollen grain was moved either upward or downward by the amplitude of the turbulence in each step. Whether the pollen grains moved upward or downward were randomly determined. The wind speed, amplitude of turbulence, and horizontal translation length (also referred to as the turbulence length) were used as inputs for the calculation. The number of repetitions of the movement of a particle in each step was determined as the quotient of the wind speed divided by the turbulence length (Fig. 5). The sampling duration was set to 180 s, taking into consideration the time required for the calculation and memory capacity of the computer. The following combinations of wind speed, turbulence amplitudes, and turbulence lengths were applied in this simulation.

Wind speed: $[0.01 \text{ m s}^{-1}, 0.5 \text{ m s}^{-1}, 1.0 \text{ m s}^{-1}, 2.0 \text{ m s}^{-1}]$.

Turbulent amplitude: $[0 \text{ m}, 0.01 \text{ m}, 0.1 \text{ m}, 0.2 \text{ m}, 0.5 \text{ m}]$.

Turbulent length: $[0.1 \text{ m}, 0.2 \text{ m}, 0.5 \text{ m}]$.

The ground level was set as -1 m , and it was assumed that once a pollen grain was deposited on the ground, the pollen grain would never be uplifted back into the air.

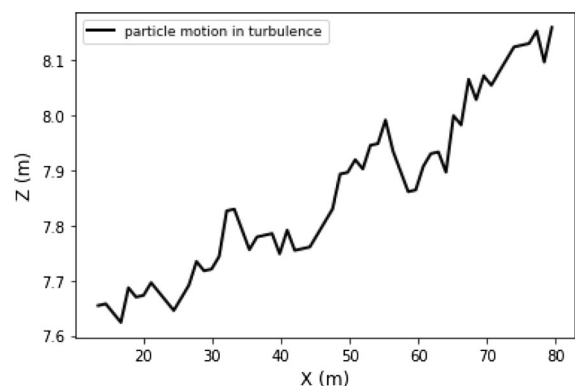


Fig. 4 Example of particle motion of in the simulation

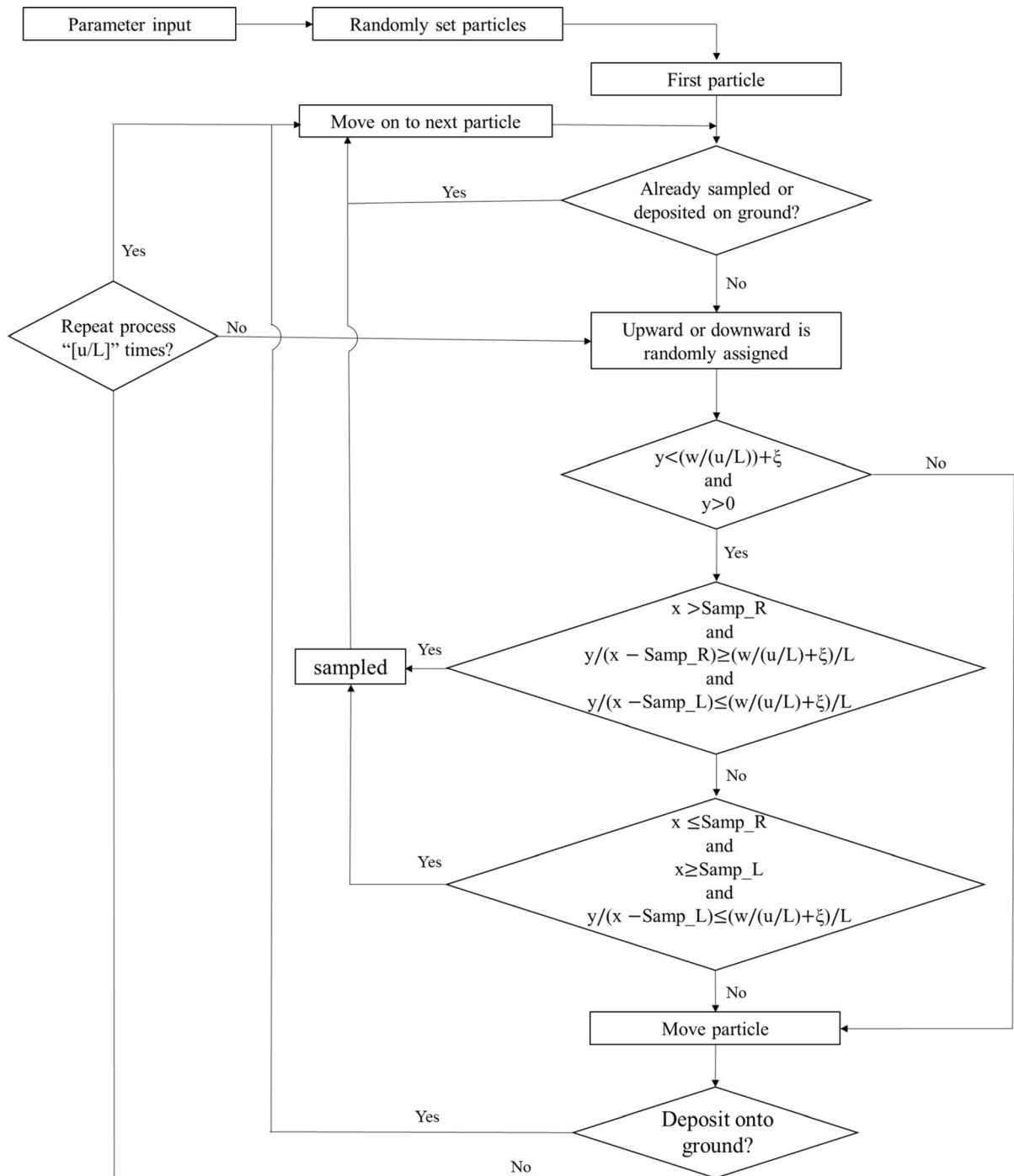


Fig. 5 Flowchart of the simulation. Samp_R and Samp_L are the x coordinate of the right edge (0.5, 0) and left edge (-0.5, 0) of the sampling plate, respectively

3.2.2 Simulation results and approximation of turbulent flow by simple vibration

For wind in a laminar flow, the number of deposited particles is expected to be calculated using Eq. (13), regardless of the wind speed.

$$\begin{aligned} N_0 &= v_s \times t \times C_0 \\ &= 1.19 \times 10^{-2} \times 180 \times 10 \\ &= 21.42(\text{grains}). \end{aligned} \quad (13)$$

The simulation results show that the number of sampled pollen grains is approximately 21 grains, as predicted for a turbulence amplitude of zero in laminar flow. Additionally, the number of sampled pollen grains increased with an increase in wind speed, which is identical to the results of the pollen sampling experiment (Fig. 6). In addition to the wind speed, the simulation shows that the number of deposited pollen grains is related to the amplitude and length of the turbulence (Fig. 7). A reasonable explanation for why these amplitudes and the turbulence length are related to the deposition is that although the direction of the vertical fluctuations is randomly determined, the turbulence can be approximated by the average waveform. When the amplitude of the turbulence was sufficiently larger than the particle deposition speed, the vertical motion was mainly dominated by fluctuation of the turbulence. Thus, the number of

pollen grains deposited on the sampling plate is proportional to the amount of air impact on the sampling plate when the pollen grains are entrained by the waveform air flow. The air motion above the sampling plate is expressed by Eq. (14), under the condition that the wind waveform is

$$F(t) = \Phi \sin\left(\frac{2\pi u}{4L}\right)t + \Psi \cos\left(\frac{2\pi u}{4L}\right)t. \quad (14)$$

Here, Φ and Ψ are related to the amplitude of the turbulence, and L is the turbulence length.

Considering that the amount of air impact on the sampling plate is proportional to the orbital distance of the vibration and that the air impact on the sampling plate only occurs for downward air motion, the number of pollen grains sampled by a passive sampler is calculated using Eq. (15), in which the length of function curve is calculated as

$$\begin{aligned} N &= k \frac{fS}{2} C \int_0^T \sqrt{1 + F(t)^2} dx \\ &= k \frac{uS}{2L} C \int_0^T \sqrt{1 + \left(\frac{\pi u}{2L} A \cos\left(\frac{\pi u}{2L}\right)t - \frac{\pi u}{2L} B \sin\left(\frac{\pi u}{2L}\right)t\right)^2} dx, \end{aligned} \quad (15)$$

where k is a constant, f is the frequency of vibration, and S is the area of the sampling plate. In this equation, the wave formula (Eq. (16)) was substituted.

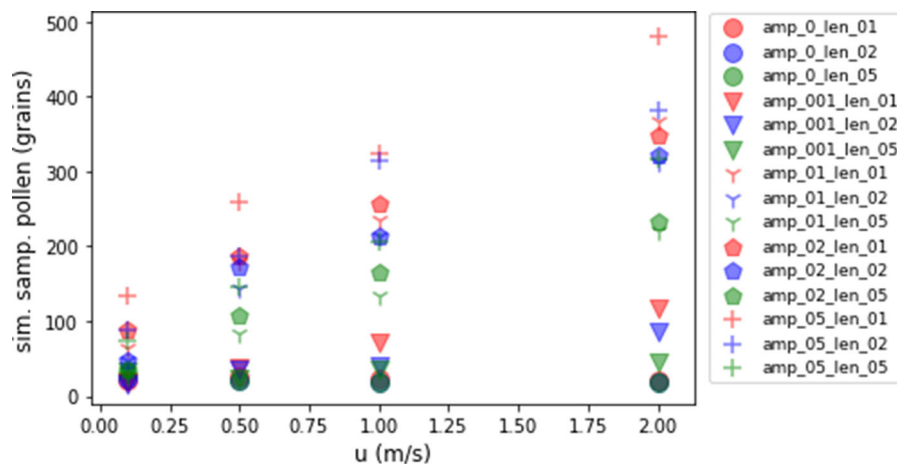


Fig. 6 Relationship between the amount of sampled pollen and wind speed (u), turbulence amplitude, and turbulence length in the simulation. [legend name] (turbulent amplitude (m), turbulent length (m)): [amp_0_len_01] (0, 0.1); [amp_0_len_02] (0, 0.2); [amp_0_len_05] (0, 0.5); [amp_001_len_01] (0.01, 0.1); [amp_001_len_02] (0.01, 0.2);

[amp_001_len_05] (0.01, 0.5); [amp_01_len_01] (0.1, 0.1); [amp_01_len_02] (0.1, 0.2); [amp_01_len_05] (0.1, 0.5); [amp_02_len_01] (0.2, 0.1); [amp_02_len_02] (0.2, 0.2); [amp_02_len_05] (0.2, 0.5); [amp_05_len_01] (0.5, 0.1); [amp_05_len_02] (0.5, 0.2); [amp_05_len_05] (0.5, 0.5)

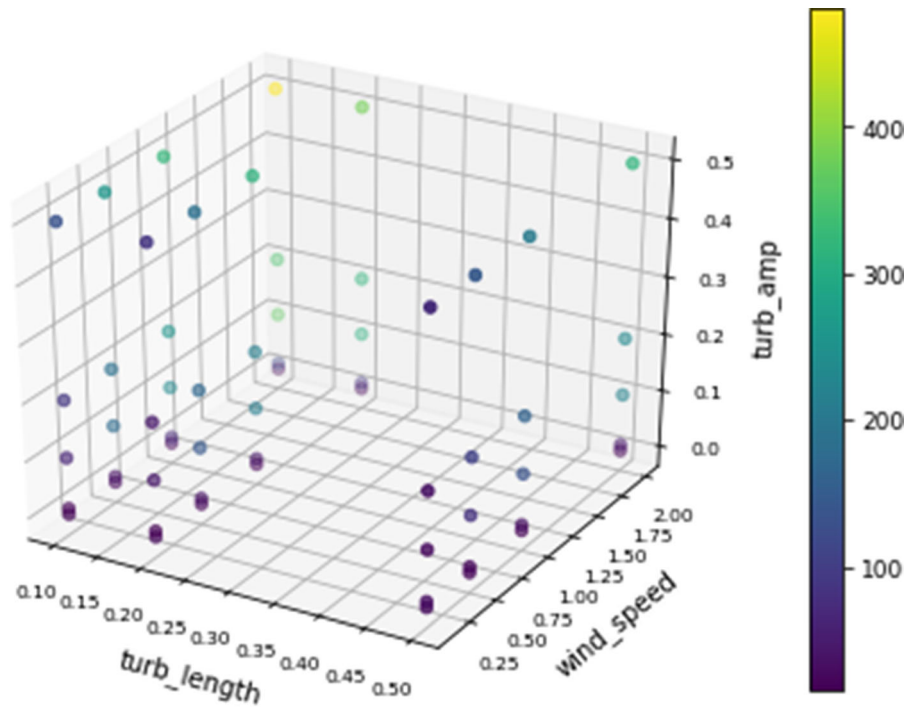


Fig. 7 Relationship between the amount of sampled pollen grains and wind speed (wind_speed), turbulence amplitude (turb_amp), and turbulence length (turb_length) in the simulation

$$f = \frac{u}{L}. \quad (16)$$

3.2.3 Evaluation of simple vibration model

To determine whether the simple vibration model is appropriate for describing the motion of the pollen grains above the sampling plate, the relationship between amount of air impact on the sampling plate and wind speed, turbulence amplitude, and turbulence length was simulated.

For simplicity, Eqs. (17) and (18) are assumed as follows:

$$\Phi = \Psi = \xi \quad (17)$$

and

$$\xi \gg v_s \quad (18)$$

where ξ is a constant.

By standardizing Eq. (15) by dividing N by N_{std} ($u = 0.1$, $L = 0.1$, $\xi = 0.01$), as in Eq. (19), the magnitude of the impact of air on the sampling plate is evaluated.

$$\frac{N}{N_{std}} = \frac{u \int_0^T \sqrt{1 + \left(\frac{\pi u}{2L} \xi \cos\left(\frac{\pi u}{2L} t\right) - \frac{\pi u}{2L} \xi \sin\left(\frac{\pi u}{2L} t\right)\right)^2} dx}{L \int_0^T \sqrt{1 + \left(\frac{\pi}{200} \cos\left(\frac{\pi}{2} t\right) - \frac{\pi}{200} \sin\left(\frac{\pi}{2} t\right)\right)^2} dx}. \quad (19)$$

Values of u from 0.1 to 2.0 in intervals of 0.5, L from 0.1 to 0.5 in intervals of 0.1, and ξ from 0.01 to 0.2 in intervals of 0.05 were substituted into Eq. (19).

From the results of the calculation (Fig. 8), the same trend as the simulation result was evident, in which larger turbulence corresponded to faster wind flow, and shorter turbulence length corresponded to an increase in the amount of air impact on the sampling plate.

Thus, the study demonstrates that the amount of pollen deposited on the passive sampler is influenced by fluctuations in the turbulent flow. In addition, even when the direction of fluctuation of the turbulence is determined randomly, pollen deposition can be described by the vibration and wave formula.

Although a simple vibration was applied in this experiment, the actual turbulence consisted of various lengths and wave amplitudes. Hence, application of the above theory to complex waves is useful for

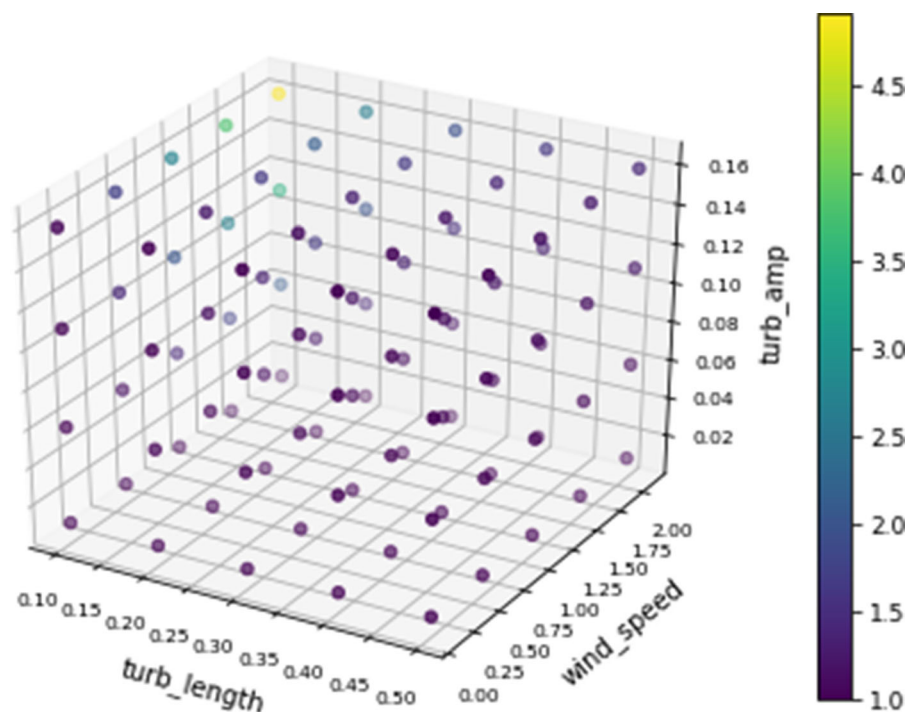


Fig. 8 Relationship between the amount of air impact on the sampling plate and wind speed (wind_speed), turbulence amplitude (turb_amp), and turbulence length (turb_length)

establishing an accurate evaluation of the airborne pollen concentration from pollen deposition data. Additionally, these experimental results are expected to be important for understanding the relationship between the distance from the pollen source and pollen deposition (Hofmann et al., 2014; Jarosz et al., 2003). Since the pollen distribution in the air and the pollen distribution bias on the sampling plate may also cause inaccurate pollen concentration evaluation (Mimić & Šikoparija, 2021), a new standard method for airborne pollen concentration evaluation is recommended for a more precise understanding of the airborne pollen distribution.

4 Conclusion

Our experimental results indicate that the commonly used correction theory does not adequately explain the relationship between gravitational sampling and volumetric sampling owing to the influence of wind speed. However, our newly proposed theory can explain the wind speed effect by considering the fluctuation of the vertical wind caused by turbulence.

The results suggest that a drastic correction is required for the determination of the airborne concentration of bioaerosols represented by pollen based on data from existing gravitational sampling methods.

Acknowledgements The authors thank Ms. Yuka Matsuda, a secretary at the Graduate School of Agriculture, Kyoto University, for her invaluable assistance throughout this study. The authors express their appreciation to the office staff at the Tokyo Institute of Technology Earth-Life Science Institute.

Funding This work was supported by JSPS KAKENHI [Grant Number 201812315].

Disclosure statement We have read and understood your journal's policies, and we believe that neither the manuscript nor the study violates any of these.

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