

# Comparability between Durham method and real-time monitoring for long-term observation of Japanese cedar (*Cryptomeria japonica*) and Japanese cypress (*Cryptomeria obtusa*) pollen counts in Niigata prefecture, Japan

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**Abstract** Japanese cedar (*Cryptomeria japonica*) pollinosis (JCP), affecting more than a quarter of the Japanese population, is a significant public health problem, due to its negative impact on daily activity. JCP patients have used the four-stage daily pollen deposition information based on the pollen monitoring over 20 years. However, the procedure for monitoring pollen was recently changed dramatically, to hourly average pollen concentration monitoring. In that type of monitoring, JCP patients cannot identify pollen exposure level because the relationship between hourly average pollen concentration and daily pollen deposition is unclear. Based on the parallel monitoring of concentration and deposition counts that we performed in Niigata prefecture, Eastern Japan, we found that the relationship between the daily pollen deposition ( $\text{pollen cm}^{-2} \text{ day}^{-1}$ ) and the daily-average pollen concentration ( $\text{pollen m}^{-3}$ ) calculated from hourly average pollen concentration was not only statistically significant but also consistent with the aerodynamic properties of pollen. Using the

relationship, we proposed new range criteria of hourly average pollen concentrations corresponding to the four stages of pollen deposition. Additionally, the conversion of pollen deposition to pollen concentration made the long-term trend analysis of the daily-average pollen concentration possible in this study area, and an increasing trend was identified at one site.

**Keywords** Japanese cedar pollen · Daily pollen deposition · Hourly average pollen concentration · Real-time pollen monitor · Pollen deposition velocity

## 1 Introduction

Japanese cedar (*Cryptomeria japonica*) pollinosis (JCP) is one of the most common types of seasonal allergic rhinitis in Japan (Saito 2014). Japanese cypress (*Cryptomeria obtusa*) pollen is another source of aeroallergens that cause allergy symptoms (Mori et al. 1999). In recent years, the number of JCP patients has markedly increased in Japan (Okubo et al. 2014). The most recent survey, which was conducted in 2008, revealed a JCP prevalence rate of 26.5% in the Japanese population (Baba and Nakae 2008). JCP is characterized by paroxysmal repetitive sneezing, watery rhinorrhea, and nasal blockage. It is frequently complicated by allergic conjunctivitis (Okubo et al. 2014). JCP has a negative impact on the performance of daily activities, work productivity, learning, sleep,

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and quality of life in people of all ages; therefore, the appearance of JCP in more than a quarter of the Japanese population presents a significant public health problem (Yamada et al. 2014). In Niigata prefecture, Eastern Japan, Japanese cedar and Japanese cypress pollen are dispersed mainly from late February or early March through April (Fujisaki 1985; Sasagawa et al. 1990). The pollen season usually lasts for about two months in Niigata prefecture, and JCP patients have allergy symptoms resulting from high-level pollen exposure during that period. The treatment of JCP varies chosen based on the severity of allergy symptoms, disease types, and the person's lifestyle (Okubo et al. 2014). Avoidance of pollen improves allergy symptoms as well as symptom-targeted treatment. It is the most effective precautionary measure and is one of the most efficacious treatments for alleviating symptoms. Therefore, it is important for JCP patients to be aware of the pollen counts on any given day and to take preventive measures in order to avoid pollen inhalation or contact (Gotoh et al. 2005).

The Niigata Prefectural Government collected data on pollen deposition from 1989 through 2011 by a passive and a gravimetric method using the Durham sampler (Durham 1946; JME 2016a). Then, the Japanese Ministry of the Environment established the *kankyo sho kafun kansoku system* (Japanese Ministry of the Environment Japanese cedar pollen observation system; Hanako-san system) (JME 2016b), an active and a volumetric method using a real-time pollen monitor, the KH-3000 (Yamato Co., Ltd., Japan) (Fukaya 2001; JME 2016c), which measures pollen concentrations, and the system has been in use nationwide since April 2008. We acquired data sets collected using the gravimetric method and the volumetric method.

In Japan, the measurement of atmospheric pollen and range criteria of pollen deposition levels were standardized in 1993 (Sahashi et al. 1993). The pollen deposition was collected daily using the Durham sampler, and it was ranked in four stages of severity (Sahashi et al. 1993). Information on pollen deposition levels was provided daily by the mass media during the pollen season and was widely used by the general public in Japan. However, the information was not sufficient for JCP patients, who wanted to know real-time pollen exposure levels. Consequently, the number of Hanako-san website users was expected to

increase, as the site displays hourly average pollen concentrations, classified into eight stages (JME 2016b). However, it is not known how the real-time pollen concentration levels listed on the Hanako-san website fit into the preexisting stages of pollen deposition classifications, even though the four-stage daily pollen deposition information is widely used in Japan.

In this study, we sought to determine hourly average pollen concentrations corresponding to the four stages of daily pollen deposition by analyzing the relationship between the two pollen counting methods. Furthermore, we characterized the long-term variation in the daily-average pollen concentration in Niigata prefecture, Japan, based on the long-term observational data.

## 2 Materials and methods

### 2.1 Locations of the sites in Niigata prefecture

Niigata prefecture is approximately 350 km north of Tokyo and is located along the Sea of Japan. The daily pollen deposition and the hourly average pollen concentration were monitored at three sites in Niigata prefecture (Fig. 1): in Niigata (Niigata Prefectural Institute of Public Health and Environmental Sciences: 37°50'N, 138°56'E, 1.5 m (the KH-3000) and 19 m (the Durham sampler) above the ground), Nagaoka (Niigata Prefectural Nagaoka Public Health Center: 37°26'N, 138°52'E, 10 m above the ground), and Joetsu (Niigata Prefectural Joetsu Public Health Center: 37°08'N, 138°14'E, 10 m above the ground).

### 2.2 Daily pollen deposition

The daily pollen deposition was monitored from February through April for 22 years (1990–2011) at Niigata and 21 years (1991–2011) at Nagaoka and Joetsu. It was measured using the Durham sampler, which involves counting the number of pollen that adheres to a white petrolatum-coated glass slide affixed to the inside of the Durham sampler over a 24-h period. The glass slide was changed at 9:00 a.m. every day. Pollen were stained with gentian violet glycerin jelly (60 mL glycerol, 10 g gelatin, 1 mL 0.1% gentian violet ethanol solution, 0.5 mL liquefied phenol, and 35 mL distilled water) and were mounted

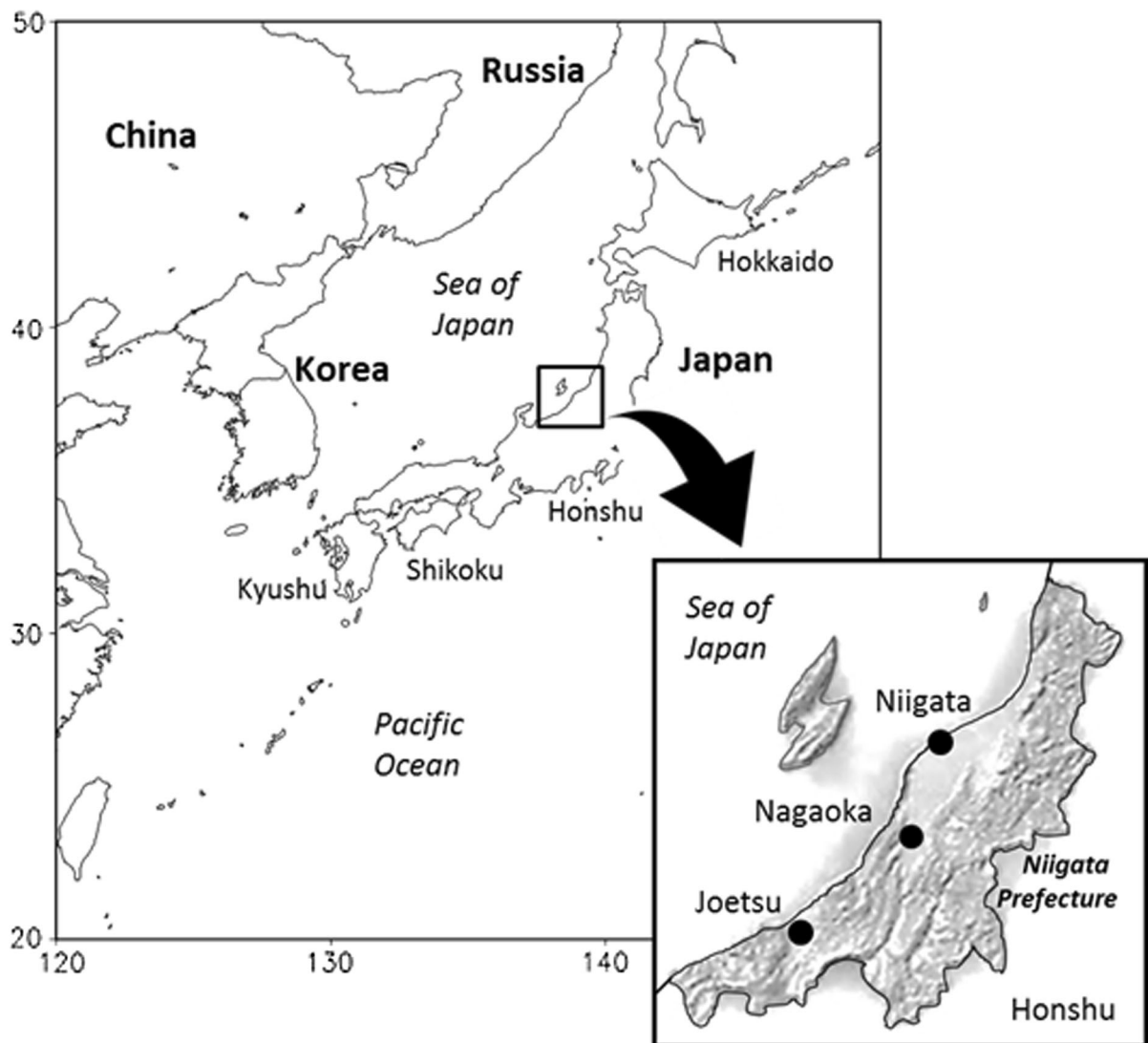


**Fig. 1** Location of study area

with a glass cover. We observed the glass slide using a light microscope and separately counted the number of Japanese cedar and Japanese cypress pollen within the glass cover (a square of 18 mm × 18 mm (3.2 cm<sup>2</sup>)). The daily pollen deposition was expressed as the total number of Japanese cedar and Japanese cypress pollen per square centimeter per day. When the glass slide was not changed due to weekends or national holidays and so forth, the pollen deposition on those days was assigned based on summarized data from a particular period.

### 2.3 Hourly average pollen concentration

The Hanako-san system has operated in Niigata prefecture from April 2008 to the present. The hourly average pollen concentration is measured using the real-time pollen monitor KH-3000 from February through April every year, and the measured values are displayed on the Hanako-san website. The KH-3000 has collected ambient air with a flow rate of 4.1 L min<sup>-1</sup> and automatically counted the number of particles with diameters of 28–35 μm, using the semiconductor laser light scattering method, because the pollen size of Japanese cedar and Japanese cypress

is approximately 30  $\mu\text{m}$ . The KH-3000 calculates the number of pollen in 60 min increments, and the results are converted to yield the number of pollen per cubic meter (Fukaya 2001).

#### 2.4 Pollen deposition velocity of Japanese cedar and Japanese cypress

Pollen released from the trees into the atmosphere causes health damage mainly on a no-precipitation day and deposits on the surface. The deposition process of a constituent in the atmosphere without precipitation is defined as dry deposition, and the deposition amount is given as the product of atmospheric concentration and deposition velocity (Pryor et al. 2008). Since Japanese cedar and Japanese cypress have similar aerodynamic particle sizes, the pollen deposition velocity ( $V_d$  ( $\text{cm s}^{-1}$ )) was calculated from the daily total pollen deposition ( $F$  (pollen  $\text{cm}^{-2} \text{ day}^{-1}$ )) and the daily-average total pollen concentration ( $C$  (pollen  $\text{m}^{-3}$ )) of the two pollen described in the previous sections by the equation;  $V_d = F/C$ .

#### 2.5 The four stages of daily pollen deposition and the eight stages of Hanako-san hourly average pollen concentration

The daily pollen deposition as measured by the Durham sampler was classified into four stages: low (0–9 pollen  $\text{cm}^{-2} \text{ day}^{-1}$ ), moderate (10–29 pollen  $\text{cm}^{-2} \text{ day}^{-1}$ ), high (30–49 pollen  $\text{cm}^{-2} \text{ day}^{-1}$ ), and very high ( $\geq 50$  pollen  $\text{cm}^{-2} \text{ day}^{-1}$ ) (Sahashi et al. 1993). The hourly average pollen concentration as measured by KH-3000 is divided into eight stages: 0, 1–19, 20–49, 50–99, 100–299, 300–499, 500–999, and  $\geq 1000$  pollen per cubic meter (JME 2016b). During the pollen season, information on pollen deposition is provided daily by the mass media and real-time hourly average pollen concentrations are displayed on the Hanako-san website.

#### 2.6 Statistical analyses

Statistical analyses in this paper were performed using the IBM SPSS Statistics 23.0 package (IBM Corp., USA). Correlation coefficients between the daily pollen deposition and the daily-average pollen concentration were evaluated using the Kendall and Spearman tests. The analysis used 10 months data

(February and March 2009–2011, April 2008–2011). For the daily-average pollen concentration, regression analysis was performed with linear regression to find trends and was evaluated using *t* tests. The analysis used 26 or 27 years data (1990 or 1991–2016). A two-tailed  $P < 0.05$  was considered to indicate statistical significance.

#### 2.7 Trajectory analysis

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLOT) model developed by the National Oceanic and Atmospheric Administrative Air Resource Laboratory, USA, was used to calculate the backward trajectory of air masses reaching Nagaoka ([http://www.arl.noss.gov/HYSPLIT\\_info.php](http://www.arl.noss.gov/HYSPLIT_info.php)) (Draxler and Hess 1997). The 5-h backward trajectories at the height of 80 m above the ground were calculated starting at the time of maximum hourly average concentration in the days that very high daily-average concentration ( $> 138$  pollen  $\text{m}^{-3}$ ) was observed in March and April 2011 in Nagaoka.

### 3 Results

#### 3.1 Relationship between daily deposition and hourly average concentration of pollen counts in Niigata prefecture

A total of 538 daily pollen deposition data (Niigata:  $n = 201$ , Nagaoka:  $n = 206$ , and Joetsu:  $n = 131$ ) were available for 10 months (February and March 2009–2011, April 2008–2011). On the same days as the daily pollen deposition, the hourly average pollen concentration was converted to the daily-average pollen concentration, starting at 9:00 a.m. The correlation between the daily-average pollen concentration and the daily pollen deposition was significant ( $r = 0.542$  by Kendall test,  $P < 0.001$ ;  $r = 0.713$  by Spearman test,  $P < 0.001$ ) (Fig. 2).

#### 3.2 Pollen deposition velocity of Japanese cedar and Japanese cypress

The pollen deposition velocity can be calculated to be  $4.2 \text{ cm s}^{-1}$  from the slope value (0.3633) of the relationship between the daily pollen deposition (pollen  $\text{cm}^{-2} \text{ day}^{-1}$ ) and the daily-average pollen

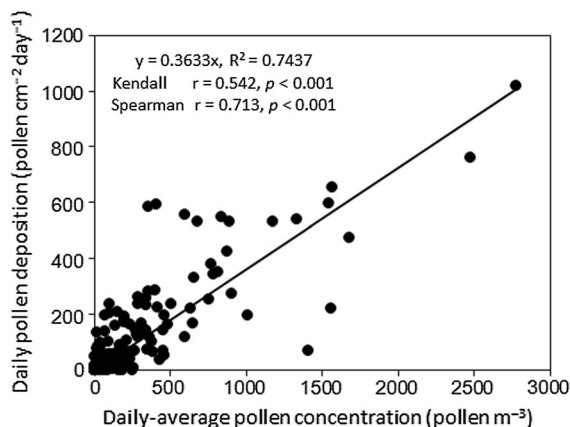


**Fig. 2** Relationship between the daily pollen deposition and the daily-average pollen concentration for 10 months (February and March 2009–2011, April 2008–2011) in Niigata prefecture ( $n = 538$ )

concentration ( $\text{pollen m}^{-3}$ ) as shown in Fig. 2 and unit conversion (coefficient: 11.57).

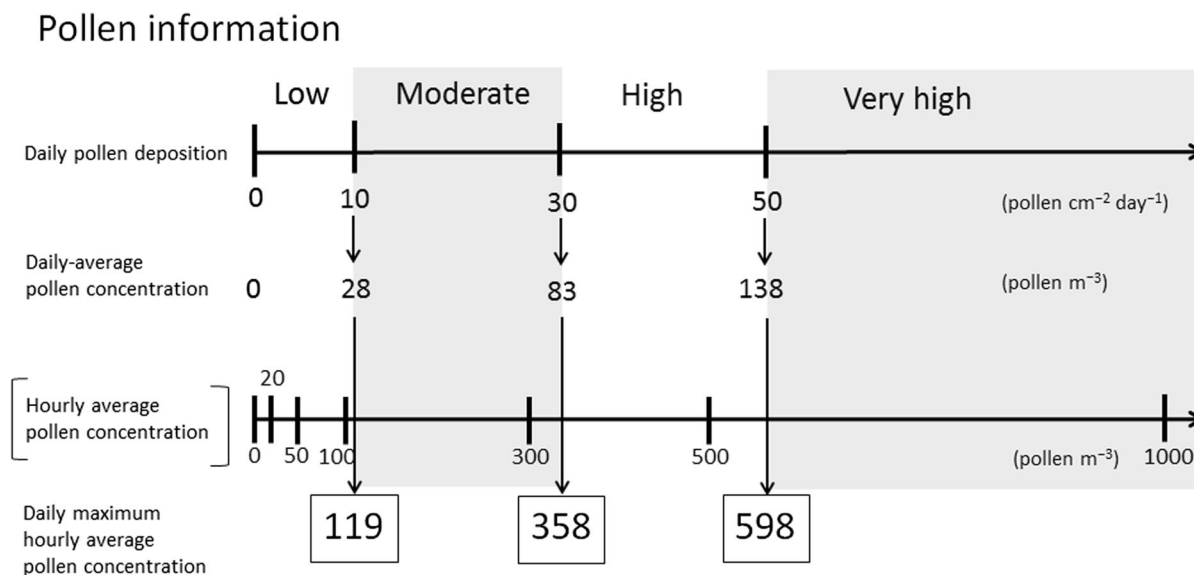
### 3.3 Thresholds for hourly average pollen concentration on the Hanako-san website based on daily pollen deposition

The thresholds for daily pollen deposition were 10, 30, and 50  $\text{pollen cm}^{-2} \text{ day}^{-1}$  (Fig. 3), based on the

Durham monitoring. Those values were converted into the daily-average pollen concentration using the slope value (0.3633) (Fig. 2) and unit conversion and were found to be 28, 83, and 138  $\text{pollen m}^{-3}$ , respectively (Fig. 3). Furthermore, we analyzed the relationship between daily-average pollen concentration ( $X$ ) and daily maximum hourly average pollen concentration ( $Y$ ) using the data from Niigata, Nagaoka, and Joetsu from 2008 through 2016 ( $n = 2782$ ). The two variables showed a significant correlation ( $Y = 4.34X$ ,  $r = 0.884$ ). The thresholds for the daily-average pollen concentration were converted into the daily maximum hourly average pollen concentration using the slope value (4.34) and found to be 119, 358, and 598  $\text{pollen m}^{-3}$ , respectively (Fig. 3).

### 3.4 Yearly variation in daily-average pollen concentrations

We analyzed the daily-average pollen concentration from Niigata for 27 years (1990–2016) and that from Nagaoka for 26 years (1991–2016). Joetsu was excluded from the trend analysis because the daily pollen deposition included many missing data. Medians of the daily-average pollen concentration in the top three were recorded in 1995, 2005, 2009, and 2011, and those of in the bottom 3 were recorded in 1992,



### Hanako-san website

**Fig. 3** Classification of the pollen exposure level in each data summation

1994, 1996, and 1999 (Table 1). The highest third quartiles of daily-average pollen concentrations at Niigata and Nagaoka were 223.4 pollen  $\text{m}^{-3}$  in 2005 and 446.2 pollen  $\text{m}^{-3}$  in 2011, respectively. Similarly, the lowest first quartiles of those at Niigata and Nagaoka were 0.7 pollen  $\text{m}^{-3}$  in 1992 and 0.0 pollen  $\text{m}^{-3}$  in 1994 and 1996, respectively (Fig. 4). Three-year moving average of the daily-average pollen concentration medians was significantly increased at Nagaoka ( $P < 0.001$ ), but very insignificantly increased at Niigata ( $P = 0.743$ ) (Fig. 5).

#### 4 Discussion

We analyzed the relationship between the daily pollen deposition and the daily-average pollen concentration to estimate a coefficient for interconversion between them. There are a few outliers in the relation, especially two outliers that are high concentration but are low deposition (Fig. 2). Those are the second outlier at Nagaoka in 2009 and 2011 (Fig. 4). In those cases, specific reasons were not confirmed such as unusual weather condition or any problems during sample collection and analysis. One of the possible reasons for overestimation of pollen concentration was that the KH-3000 measured not only Japanese cedar and Japanese cypress pollen grains but particles of 28–35  $\mu\text{m}$  size as well. Although our data set included those outliers, the daily-average pollen concentration was significantly correlated with the daily pollen deposition ( $P < 0.001$ ) (Fig. 2). Furthermore, from the slope of the relationship between them, the pollen deposition velocity was estimated to be 4.2  $\text{cm s}^{-1}$ . The aerodynamic diameter ( $D$  ( $\mu\text{m}$ )) of atmospheric particles can be calculated from the actual particle diameter ( $d$  ( $\mu\text{m}$ )) and its density ( $\rho$  ( $\text{g cm}^{-3}$ )) by the equation:  $D = \sqrt{\rho * d}$  (Vallero 2007). When the actual particle diameter of Japanese cedar and Japanese cypress pollen grains ranges 28–35  $\mu\text{m}$ ,

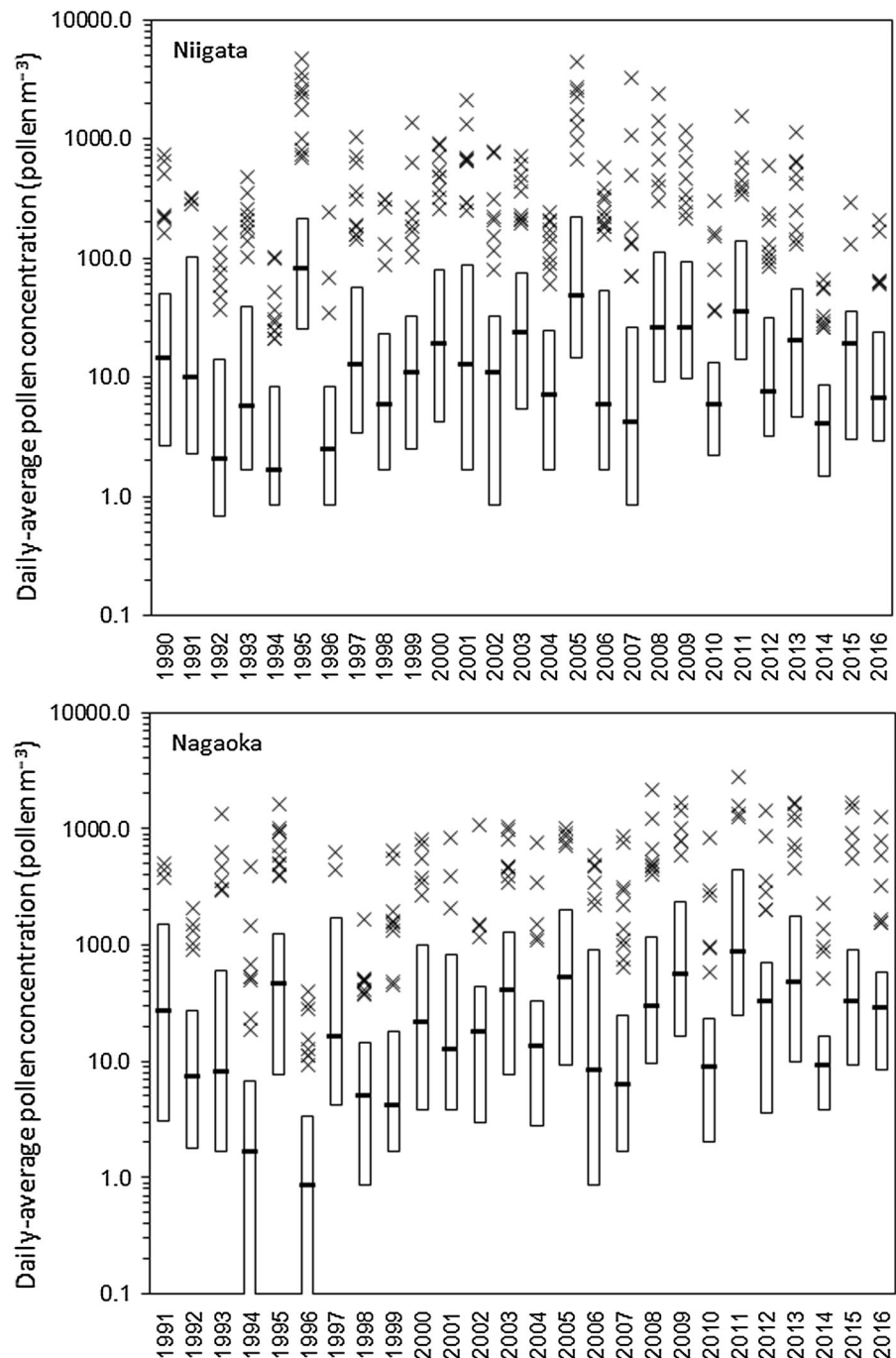
their aerodynamic diameters are estimated to range 25–36  $\mu\text{m}$ , because their particle densities have been found to be  $1.06 \pm 0.08 \text{ g cm}^{-3}$  and  $0.82 \pm 0.04 \text{ g cm}^{-3}$  for Japanese cedar and Japanese cypress, respectively (Hirose and Osada 2016). The theoretical deposition velocity against the aerodynamic diameter of atmospheric particles has been reported (Pryor et al. 2008). In that report, the average deposition velocity of particles with aerodynamic diameters of 25–36  $\mu\text{m}$  was estimated to be approximately 5–7  $\text{cm s}^{-1}$ . The deposition velocity is influenced by particle size, weather conditions, and the collection properties of the surface. The pollen size and the adhesive glass slide surface were uniform, but weather conditions varied. Taking those variables into account, the deposition velocity estimated from the atmospheric concentration and deposition of the pollen was considered to be consistent with the theoretical deposition velocity of particles of that diameter. Thus, we provided evidence that the relationship between the pollen counts is not only statistically significant but also fits the aerodynamic property of pollen. The comparison of the daily pollen deposition, as measured by the Durham sampler, and the hourly average pollen concentration, as measured by the KH-3000, was carried out in Japan (e.g., Yamagata prefecture (Takahashi et al. 2001), Wakayama prefecture (Seno et al. 2003), and the Tokyo metropolitan area (Imai et al. 2005)). Those reports found that the hourly average pollen concentration correlated well with the daily pollen deposition ( $r = 0.65\text{--}0.966$ ), and the KH-3000 was useful as a real-time pollen monitor, but the reliability of the relationship between the two pollen counts was not validated by the aerodynamic property of pollen such as the pollen deposition velocity evaluated in this study.

The Durham monitoring requires the time and labor of skilled technicians, who must be able to differentiate pollen grains by their shape under a light

**Table 1** Medians of the daily-average pollen concentration in the top 3 and the bottom 3 at the Niigata (1990–2016) and Nagaoka (1991–2016) sites

| Site                                    | Medians in the top 3 |             |             | Medians in the bottom 3 |            |            |
|-----------------------------------------|----------------------|-------------|-------------|-------------------------|------------|------------|
| Niigata (pollen $\text{m}^{-3}$ , year) | 82.4 (1995)          | 49.3 (2005) | 36.2 (2011) | 1.7 (1994)              | 2.1 (1992) | 2.6 (1996) |
| Nagaoka (pollen $\text{m}^{-3}$ , year) | 87.7 (2011)          | 56.6 (2009) | 53.5 (2005) | 0.8 (1996)              | 1.7 (1994) | 4.2 (1999) |

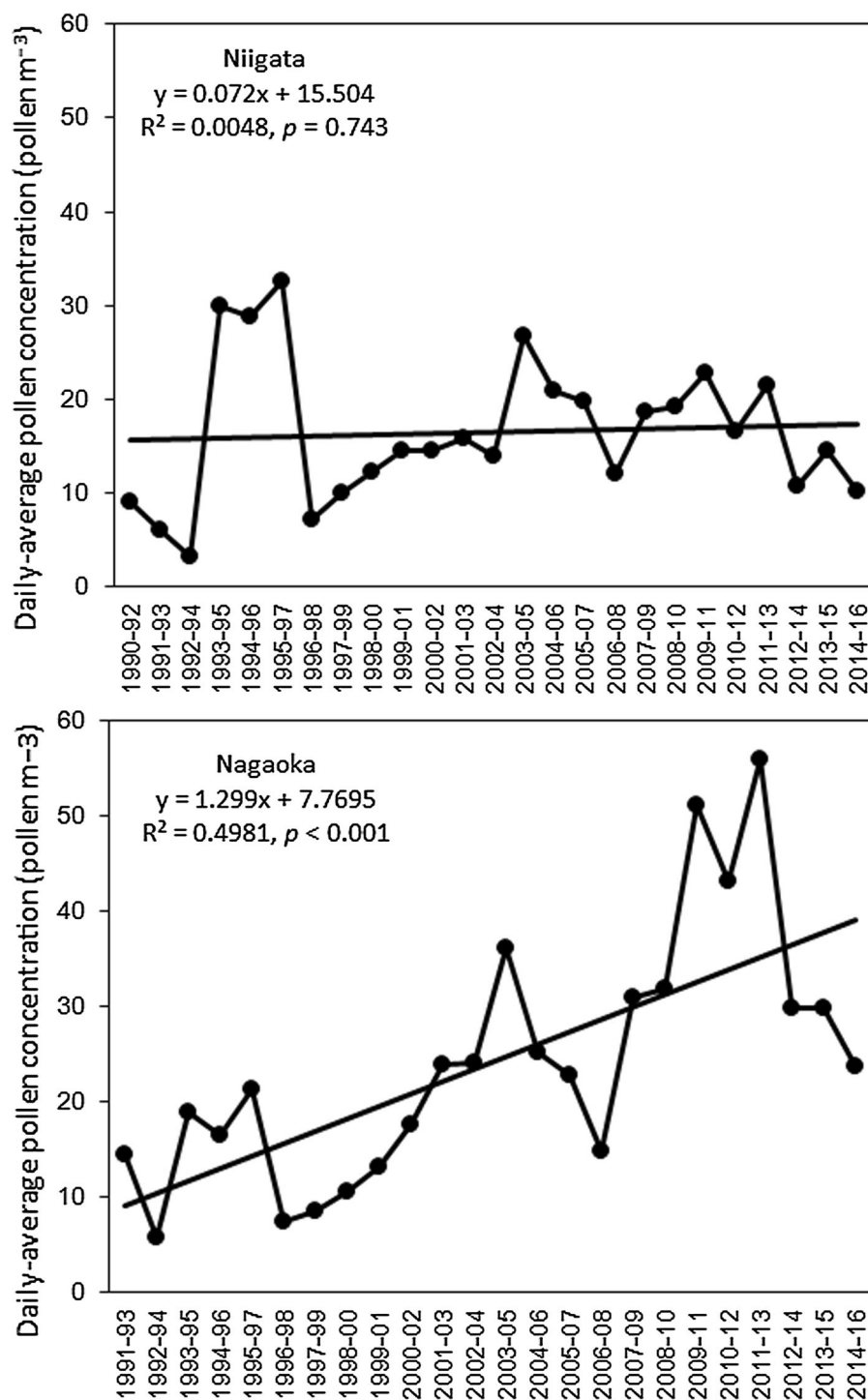
**Fig. 4** Yearly variation in the daily-average pollen concentration in March and April at the Niigata and Nagaoka sites. Bold horizon bars in rectangular boxes are medians. Rectangle tops and bottoms are the third and first quartiles, respectively. Boxes enclose the first through third interquartile range. Crosses show outliers, characterized as data that are more than 1.5 interquartile ranges from the third quartile



microscope. Conversely, the hourly average pollen concentrations are determined automatically by the KH-3000 and to find on the Hanako-san website. Despite the KH-3000 counting particles other than Japanese cedar and Japanese cypress pollen, the daily-average pollen concentration was significantly

correlated with the daily pollen deposition. Thus, we confirmed that the KH-3000 provided very efficacious data (i.e., hourly average pollen concentrations). Therefore, new range criteria are required for JCP patients to use hourly average pollen concentrations listed on the Hanako-san website in the same way as

**Fig. 5** Three-year moving average of the daily-average pollen concentration medians at the Niigata and Nagaoka sites



daily pollen deposition. We set new criteria for evaluation of the hourly average pollen concentration, which is automatically monitored by the KH-3000 because hourly average pollen concentrations listed on

the Hanako-san website are an effective tool. As this indicator aims to encourage residents to pay attention to their pollen exposure, we proposed that the daily maximum hourly average pollen concentration is a

new indicator as follows: low (0–118 pollen  $\text{m}^{-3}$ ), moderate (119–357 pollen  $\text{m}^{-3}$ ), high (358–597 pollen  $\text{m}^{-3}$ ), and very high ( $\geq 598$  pollen  $\text{m}^{-3}$ ) (Fig. 3). When a JCP patient wants to know the pollen exposure level on a particular day at a particular time, she/he can determine the provisional pollen exposure level of the day by applying the maximum hourly average pollen concentration recorded up to the time of the day to the above-mentioned new criteria.

The thresholds for hourly average pollen concentrations in Niigata were proposed by statistical analysis of monitoring data in order to be more effective for JCP patients. However, it was considered insufficient for use as medical information because the thresholds for hourly average pollen concentrations were determined based on range criteria of the daily pollen deposition. Therefore, novel criteria should be defined by the relationship between the hourly average pollen concentration and exacerbation of allergy symptoms. JCP is still poorly understood. The relationships between real-time pollen concentration levels, exposure time to pollen, amount of pollen in the nasal cavity, and the variation in allergy symptoms and symptomatic degree, and so forth need to be clarified in order to better understand JCP. Personal pollen exposure has been monitored using the personal aeroallergen sampler (PAAS) (Yamamoto et al. 2015, 2007), a passive sampler for aeroallergens and personal pollen sampling (Yamamoto et al. 2007). The research is promising and may lead to a better understanding of JCP. Moreover, the development of the next-generation pollen information, namely personal pollen information, was reported from Austria (Kmenta et al. 2014). The system includes an innovative forecasting model that incorporates bi-hourly pollen data, traditional pollen forecasts, meteorological data, and recent symptom data and calculates the personal symptom load in real time. In the next phase of research, we hope that personal pollen information will be developed in Japan and that a new function for individuals may be added to the Hanako-san system.

Long-term variation in daily-average pollen concentrations was analyzed to elucidate the features of the yearly pollen dispersion in Niigata. On comparing the medians of daily-average pollen concentrations between Niigata and Nagaoka, we found that although the chronological order was slightly different, from one site to another, there were no striking differences in the yearly variation of the medians. On the highest

and lowest quartiles of daily-average pollen concentrations, those data in Nagaoka fluctuated more than those in Niigata. The widespread Japanese cedar reforestation took place throughout Japan after World War II for flood control and forest conservation, as well as to secure building materials. The reforested trees reached the age of 30 years in the latter half of the 1960s, and their pollen-producing capacity was felt. The area of the reforested trees increased approximately 2.2 times in Japan, from 1.77 million ha in 1990 to 3.97 million ha in 2012 (JMAFF 2012). Although it is easy to assume that amount of the pollen produced would increase from year to year as the reforested area increases, the pollen count fluctuates greatly from year to year (Fig. 4), because pollen output is strongly influenced by the weather conditions of the last year. However, in terms of long-term variation, the pollen count at Nagaoka is considered to reflect pollen production trends, but not at Niigata (Fig. 5). Niigata prefecture extends north and south and faces the Sea of Japan on the west (Fig. 1). The mountainous areas lie east and south of Niigata prefecture. The Niigata site is located nearer the coastline (about 3 km inland) than the Nagaoka site, and the mountainous areas are distant, at more than 20 km beyond the vast Niigata Plain. The Nagaoka site is substantially closer to the mountainous areas than the Niigata site. The Japan Meteorological Agency provides meteorological data from the meteorological observatories in Japan, based on 30 years of observation. The database shows that south-southwest (winds from the mountainous area) is the dominant wind direction in March and April in Nagaoka, and west and west-southwest (winds from the Sea of Japan) are dominant in March and April in Niigata (JMA 2017). Thus, it is easy to see that the influence of forested areas would be felt more frequently in Nagaoka than Niigata in that season. Therefore, it is understandable that the three-year moving average of the daily-average pollen concentration medians at Nagaoka showed a significant increasing trend ( $P < 0.001$ ) (Fig. 5).

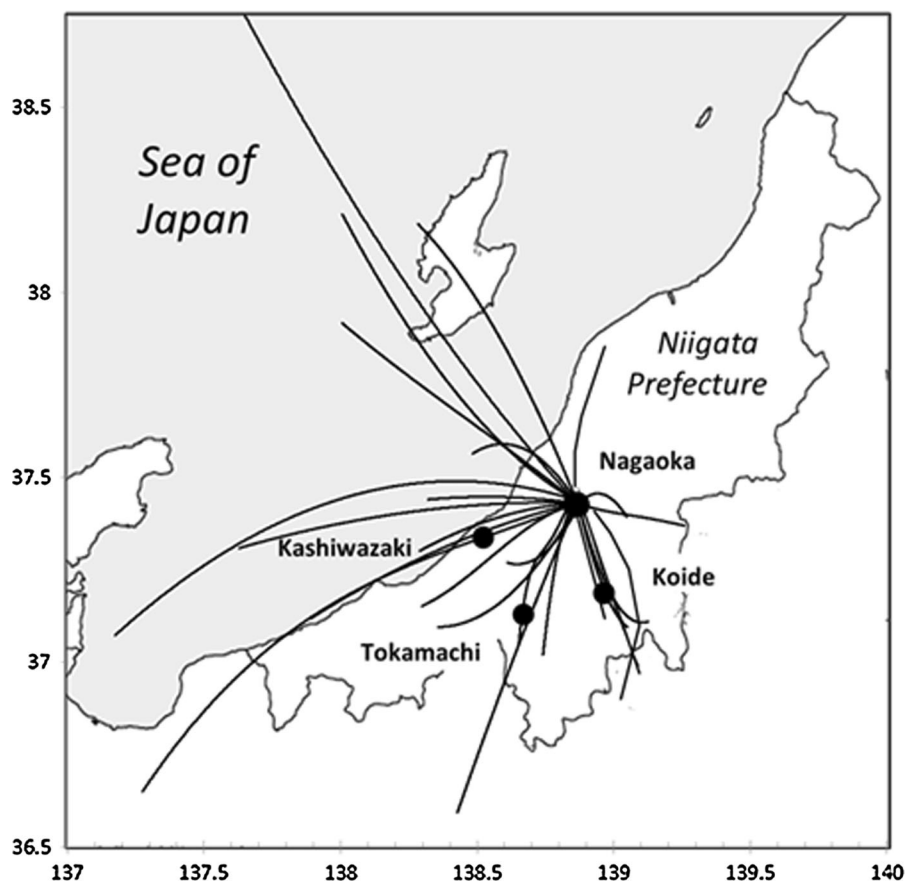
In addition, we conducted air mass trajectory analysis (Draxler and Hess 1997) to investigate the source area of pollen to Nagaoka and obtained interested results. The data of 2011 were analyzed as a typical example, because the median of daily-average pollen concentration in 2011 was the highest in the long-term trend (Fig. 4). We drew 5-h backward

trajectories starting from the time when daily maximum concentrations were recorded by using the data of 27 days that very high daily-average pollen concentrations ( $> 138 \text{ pollen m}^{-3}$ ) were observed in 2011 (Fig. 6). It was obvious that the air mass streamlines flow into Nagaoka from the southeast to the west, except for some cases, and Japanese cedar and Japanese cypress that were planted in those areas are the main sources of pollen observed in Nagaoka. Furthermore, we analyzed the relationships between pollen concentration at Nagaoka and meteorological observation data in pollen source areas (Koide, Tokamachi, and Kashiwazaki shown in Fig. 6). Those analyses investigated the correlation between medians of the daily-average pollen concentration at Nagaoka (March and April: 1991–2016) and the meteorological data (total precipitation, average temperature, and total sunshine duration) at the three sites in the previous summer (July and August: 1990–2015) (JMA 2017). As a result, the pollen concentration at Nagaoka showed significant correlation with the meteorological

parameters at the three sites (by Spearman test,  $n = 26$ ), negative in precipitation (Koide:  $r = -0.464$ ,  $P < 0.05$ ; Tokamachi:  $r = -0.400$ ,  $P < 0.05$ ; Kashiwazaki:  $r = -0.505$ ,  $P < 0.01$ ), positive in temperature (Koide:  $r = 0.624$ ,  $P < 0.01$ ; Tokamachi:  $r = 0.576$ ,  $P < 0.01$ ; Kashiwazaki:  $r = 0.569$ ,  $P < 0.01$ ), and positive in sunshine duration (Koide:  $r = 0.713$ ,  $P < 0.001$ ; Tokamachi:  $r = 0.656$ ,  $P < 0.001$ ; Kashiwazaki:  $r = 0.586$ ,  $P < 0.01$ ). These results suggest that the increase in pollen concentration at Nagaoka may be affected by decreasing precipitation, rise in temperature, and increasing sunshine duration in the pollen source areas.

In conclusion, we provide evidence that the relationship between daily pollen deposition and daily-average pollen concentration is not only statistically significant but also accounts for the aerodynamic property of pollen. This study is the first to provide thresholds for the use of hourly average pollen concentrations to limit personal exposure in Niigata.

**Fig. 6** Air mass trajectories of the days that very high daily-average concentrations ( $> 138 \text{ pollen m}^{-3}$ ) were observed in March and April in 2011 in Nagaoka. For 27 days, 5-h backward trajectories were analyzed starting at the time of maximum hourly average concentration of the day and the height of 80 m above the ground



Furthermore, differences in long-term trends were found in the three-year moving average of the daily-average pollen concentration medians, depending on the site. Further studies will be required to define novel range criteria for hourly average pollen concentration in Japan and to elucidate the detailed mechanisms of JCP with additional pollen monitoring and clinical studies.

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