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Title: High temporal resolution of airborne Ambrosia pollen measurements above the source reveals emission characteristics

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Abstract: The aim of this study was to setup a field experiment that will collect suitable data for parameterizing the release of Ambrosia pollen. The study took place on the Pannonian plain during three flowering seasons: 2014, 2015 and 2016. The sampling of airborne pollen was performed using volumetric spore traps at 0.5 m and 5 m above the canopy level of a strong homogenous Ambrosia artemisiifolia L. field (10 m × 10 m) in three temporal resolutions: 1 h, 7.5 minutes and 1.07 minutes. This high temporal resolution revealed characteristics that were hitherto unknown. Pollen production per day was estimated to range from 6.38 billion to 770 billion pollen grains for the whole experimental field. Measurements of meteorological parameters included: temperature, relative humidity, solar radiation, precipitation, wind speed and wind direction. Weak correlations between pollen concentration and meteorological parameters were obtained, indicating nonlinear relationships among them. The distribution of pollen concentration with respect to meteorological parameters showed that high pollen concentrations coincide with temperature in the interval 20-24°C, and that relative humidity of around 95% can delay or even switch off the emission, while the turbulent kinetic energy of a wind less than $0.1 \text{ m}^2 \text{ s}^{-2}$ is sufficient for lifting the pollen into the air. The test of the new model for Ambrosia pollen emission processes showed a strong correlation between measured and modeled data when the estimated emission capacity for the field was in the range of 45-75 billion pollen grains per day.

ATMENV-D-18-00419R1 High temporal resolution of airborne *Ambrosia* pollen measurements above the source reveals emission characteristics

Reviewer #1: I appreciate the work of the authors to elaborate this new version, which I find much improved and highly interesting. I have very few comments now that I summarize here:

Reply: The authors thank to the reviewer for helping us to improve the quality of the paper.

Reviewer #1: l. 49-50, I do not know the policy of Atmospheric environment with regard to Keywords but I would suggest not use in them words already appearing in the title. So, I propose to consider as Keyword Ragweed (instead of Ambrosia) and pollen production (added or instead pollen emission).

Reply: There is no strict rule regarding the words used in the title and as Keywords in Atmospheric Environment. In the new version we have added “ragweed” instead of Ambrosia, however we kept “pollen emission” since the emission process is in the focus of the paper.

Reviewer #1: l. 163, consider to add "of" between "in the source area and" and "daily rate"

Reply: It is added in the new version.

Reviewer #1: l. 188, consider to add "the" between "During" and "entire"

Reply: It is added in the new version.

Reviewer #1: l. 188, consider to add "," after "period"

Reply: It is added in the new version.

Reviewer #1: l. 203, consider to add "," after "A. artemisiifolia plants"

Reply: It is added in the new version.

Reviewer #1: l. 208, consider to move "(" from before "in case" to before "i.e. turbulence"

Reply: It is moved in the new version.

Reviewer #1: l. 209, consider to add "," between "while" and "in case" and again "," after "velocities"

Reply: It is added in the new version.

Reviewer #1: l. 209-210, change from "together with Lanzoni traps is expected to give information on background levels of A. artemisiifolia airborne pollen concentrations." To "it is expected to give information on background levels of A. artemisiifolia airborne pollen concentrations together with Lanzoni traps."

Reply: It is changed in the new version.

Reviewer #1: l. 212, consider to add "," after "3 h"

Reply: It is added in the new version.

Reviewer #1: l. 235, consider to add "," between "concentrations" and "a meteorological"

Reply: It is added in the new version.

Reviewer #1: l. 548, consider to add "Ambrosia" in the title

Reply: It is added in the new version.

Reviewer #1: l. 564, add "I" at the end of "Rantio-Lehtimäk"

Reply: It is added in the new version.

Reviewer #1: l. 662, consider to add "of 10x10 m² surface" after "A. artemissiifolia"

Reply: It is added in the new version.

Reviewer #1: l. 664, Is it correct or precise enough to talk about "sampling frequency"? What about using "resolution" instead of "frequency"?

Reply: It is corrected to “resolution” in the new version.

Reviewer #1: l. 679, I think that "and" between "parameter" and "in the future" is not needed. I propose to eliminate it.

Reply: It is eliminated in the new version.

**High temporal resolution of airborne *Ambrosia* pollen measurements above the source reveals
emission characteristics**

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Abstract

The aim of this study was to setup a field experiment that will collect suitable data for parameterizing the release of *Ambrosia* pollen. The study took place on the Pannonian plain during three flowering seasons: 2014, 2015 and 2016. The sampling of airborne pollen was performed using volumetric spore traps at 0.5 m and 5 m above the canopy level of a strong homogenous *Ambrosia artemisiifolia* L. field (10 m × 10 m) in three temporal resolutions: 1 h, 7.5 minutes and 1.07 minutes. This high temporal resolution revealed characteristics that were hitherto unknown. Pollen production per day was estimated to range from 6.38 billion to 770 billion pollen grains for the whole experimental field. Measurements of meteorological parameters included: temperature, relative humidity, solar radiation, precipitation, wind speed and wind direction. Weak correlations between pollen concentration and meteorological parameters were obtained, indicating nonlinear relationships among them. The distribution of pollen concentration with respect to meteorological parameters showed that high pollen concentrations coincide with temperature in the interval 20-24°C, and that relative humidity of around 95% can delay or even switch off the emission, while the turbulent kinetic energy of a wind less than $0.1 \text{ m}^2 \text{ s}^{-2}$ is sufficient for lifting the pollen into the air. The test of the new model for *Ambrosia* pollen emission processes showed a strong correlation between measured and modeled data when the estimated emission capacity for the field was in the range of 45-75 billion pollen grains per day.

Keywords: aerobiology; ragweed; measurements; pollen emission; meteorological conditions; turbulent kinetic energy

60 1. Introduction

61

62 Airborne pollen is considered as the main trigger of allergic diseases that affects as many as 15
63 million people in Europe (Bousquet, 2013). Avoidance of the exposure to airborne pollen is an
64 important part of managing allergic symptoms (Peden and Reed, 2010). Effective allergy
65 management requires the support of measurements and models that can provide patients and
66 medical doctors with temporally resolved information on the quantity of allergenic pollen
67 suspended in the atmosphere. This quantity is known to vary with climate, meteorological
68 conditions, geography and vegetation (Scheifinger et al., 2013) but also with the location of the
69 measurement system with respect to the distance from the emission source (Raynor et al., 1968).

70 Forecasting airborne pollen concentrations relies on the analysis of relationships between
71 airborne pollen concentrations and various environmental variables (Scheifinger et al., 2013).
72 Whatever method is used to explore relationships (e.g. regression analysis or more elaborate
73 machine learning techniques) the analysis should be performed on datasets able to describe the full
74 variability of the analysed phenomena. Sofiev et al. (2006) theoretically argued that if proper
75 emission fluxes could be developed, the atmospheric models could provide reliable estimates of the
76 atmospheric transport of pollen. To achieve this goal, detailed measurements of the exact emission
77 from the plants are required.

78 The Hirst-type volumetric method (Hirst, 1952) is commonly used for measuring
79 atmospheric concentrations of allergenic pollen. The standardized methodology suggests that the
80 sampling should be performed at the roof level to avoid the influence of local sources (Galán et al.,
81 2014). Many studies have shown that roof-level measurements underestimate concentrations near
82 the source and this is particularly pronounced for airborne pollen coming from herbaceous plants
83 (Alcázar and Comtois, 2000; Rantio-Lehtimäki et al., 1991; Raynor et al., 1973; Spieksma et al.,
84 2000). The further pollen is transported from the source, mixing within the lower atmospheric
85 boundary layer is more efficient. This leads to dilution of emitted concentrations (Chamecki and
86 Meneveau, 2011; Sofiev, 2017).

87 Devices used for sampling in operational aerobiological networks (e.g. Lanzoni VPPS,
88 Burkard Spore Trap, Burkard Scientific SporeWatch Sampler) are constructed to continuously
89 sample a known volume of air through an orifice and deposit the suspended particles on a sticky
90 surface. The speed at which the sampling surface moves behind the orifice determines the temporal
91 resolution of the collected dataset. To enable continuous work for several days, the speed of rotation
92 of the sampling surface is usually set to 2 mm h^{-1} which, with the standard sampling orifice 14 mm
93 $\times 2 \text{ mm}$, limits the finest temporal resolution to 1 h (BAF, 1995).

Such a coarse temporal resolution is sufficient to describe seasonal patterns, while monitoring at roof-level reflects the regional variability that includes both the emission and the atmospheric transport. However, for detailed insights into emission processes, higher temporal resolution is required for the detection of variability in the immediate vicinity of the source.

Common ragweed (*Ambrosia artemisiifolia* L.) is a noxious invasive species introduced to Europe from North America. One of the main centres of its European distribution is the Pannonian Plain which is proved to be an important source of aeroallergens throughout Europe (Smith et al., 2013). Common ragweed is often found in farmlands. It is a source of highly allergenic pollen, which is reported to induce asthma more often than other pollen types (Dahl et al., 1999). *A. artemisiifolia* L. shows clear diurnal periodicity with respect to the opening of male flowers and the release of pollen that corresponds to the increase of temperature and the decrease of relative humidity (Bianchi et al., 1959). However, threshold values for flowering were observed in the laboratory conditions and real-life tests in the open field are required to confirm their suitability for mechanistic parameterization of the emission process. Two major field experiments were conducted in the USA to provide data needed for exploring the dispersion and deposition of ragweed pollen grains (Martin et al., 2009; Raynor et al., 1970). These setups successfully described the temporal distribution of concentration peaks recorded above the source (Ogden et al., 1969), as well as the biological aspect of pollen maturation, presentation and release (Martin et al., 2009; 2010). All these mentioned studies showed that pollen emission processes are complicated and require measurements in high temporal resolution to capture the variability and the conditions in the field. In addition, none of these studies sampled pollen at the sufficient temporal resolution to enable more precise assessment of emission patterns and flux, which is needed for parameterization in dispersion models (Prank et al., 2013). Finally, all previous studies of ragweed pollen emission have been conducted in North America which is the native range of *A. artemisiifolia* L. distribution and the obtained relationships need to be confirmed with measurements in its invasive range as suggested by Ogden et al. (1969), who assumed that the location with different meteorological conditions could lead to shifts in the emission.

The first aim of this study was to setup a field experiment to collect suitable data for the characterization of *A. artemisiifolia* airborne pollen release from the source and the influence of meteorological conditions. Secondly, the collected samples are expected to show if a more temporally detailed analysis brings additional information about the emission processes.

2. Materials and methods

2.1. Location of the study area and *Ambrosia* pollen source characteristics

The study was conducted in Backi Maglic (45°21'46.3" N, 19°31'55.9" E), situated in the Pannonian part of Serbia (Fig. 1A) which is known as being heavily invaded by *Ambrosia* plants (Skjøth et al., 2010). The local source of *Ambrosia* airborne pollen was created by covering a 10 m × 10 m patch with soil from a heavily infested agriculture area. This introduced a notable seed pool and resulted in growth of numerous ragweed plants. After the plants emerged, the source area was modified to contain 500 equally distributed *Ambrosia* plants, which allowed for full development of the plant habitus and thus lead to full pollen emission potential. An additional effort was made to keep the homogenous *Ambrosia* population free from other weeds. This procedure has been repeated in 2014, 2015 and 2016.

Heavily infested backyards in populated areas show similar environments and thus the source area prepared represents the real exposure risk of inhabitants to airborne ragweed pollen. In order to ensure that the created local source area dominates in airborne pollen samples, all ragweed plants were removed within the range of at least 500 m around the source. The given buffer is considered sufficient for sensors measuring at 0.5 m and 5.5 m above the canopy (Burba and Anderson, 2005) and also is in line with findings that only 5% of ragweed pollen is transferred further then 100m from the source area (Raynor et al., 1970).

The number and percentage of plants that release pollen were recorded on the experimental field daily, to determine the time of the full pollen release. In all three seasons all plants started to release pollen by 15th August. Each day starting from this date, between one and three male flowers per inflorescence matured and released pollen.

In order to determine the strength of our pollen source we estimated pollen production per plant following the methodology described in Fumanal et al. (2007). Five fully developed plants from different locations of the source were chosen. For each plant we counted the number of: racemes (nR), male heads in five representative racemes (nMH) and male flowers in five representative male heads (nMF). Finally, total of 15 closed male flowers were extracted (five from each chosen plant) and macerated separately in 100 microliters of distilled H₂O. Complete suspension of pollen from each flower is transferred to a microscopic slide. The complete surface of the slide was analysed using light microscope (×200 magnification) and all pollen grains were counted to get the number of pollen grains per flower (nPG). The total pollen production per plant was computed from the measured parameters ($P = nR \times nMH \times nMF \times nPG$) and we presented both

162 the average so as the maximum and minimum value. Apart from total pollen production per plant,
163 knowing the number of plants in the source area and of daily rate male flowers are opened (1-3 per
164 male head) we were able to estimate range of daily pollen production of the pollen source area
165 which is required input for the emission model. The lower limit is obtained by multiplying number
166 of plants in source area with average pollen production per plant assuming only one male flower is
167 opened per male head and the upper limit is obtained by multiplying number of plants in source
168 area with average pollen production per plant assuming three male flowers are opened per male
169 head.

170 On 2nd September 2015 (18:00-20:00) the source was removed to confirm that the majority
171 of pollen recorded above the source was coming from the plants beneath. Also, on 1st September
172 2016 (9:00-10:00), half of the source was removed (central square 7 m × 7 m) to explore the effect
173 of the source size on pollen records (Supplementary material Fig. S1). The management of the
174 source shook plants and resulted in a notable artificial pollen release. Although these periods give
175 estimates of the potential exposure for workers that eradicate ragweed during flowering, we
176 removed these data when analysing normal emission patterns and relationships with meteorological
177 data.

178

Fig. 1. (A) Location of the regional Hirst-type pollen samplers in relation to study area and (B) Digital Surface Model depicting the height of objects surrounding the artificial *Ambrosia* pollen source in Backi Maglic and location of meteorological station and Burkard Scientific SporeWatch samplers in the center (asterisk) so as Lanzoni VPPS2010 samplers (filled triangle, filled circle and filled square) installed at 1.5 m above the ground in close vicinity of the pollen source.

179 2.2. Airborne pollen data

180

181 Simultaneous sampling of airborne pollen and meteorological parameters was performed in the
182 centre of the artificially created *Ambrosia* pollen source during three flowering seasons: 21th August
183 – 12th September 2014, 16th July – 5th September 2015 and 29th July – 8th September 2016. In
184 addition to measurements in the centre of the source, to serve as control for background levels,
185 pollen concentrations were recorded at 1.5 m above ground level in close vicinity of the source
186 towards dominant wind directions (20 m towards North-West in 2015, 15 m towards East in 2015
187 and 60 m towards East in 2015 and 2016). During the entire study period, airborne *Ambrosia* pollen
188 was recorded also at regional aerobiological stations situated on the roof-level in Sombor, Vrbas
189 and Novi Sad (Fig. 1A).

190 The sampling of airborne pollen was performed using volumetric spore traps of the Hirst
191 design (Hirst, 1952). Air is sucked into the trap at a rate of 10 l min⁻¹ through a 2 mm × 14 mm
192 orifice. Behind the orifice the air flows over a slowly rotating drum that moves past the inlet and is
193 covered with an adhesive coated, transparent plastic tape. Particles in the air land on the tape to give
194 a time-related sample (Emberlin, 2000), while the relationship between the rotation speed and the
195 width of the orifice determines the maximal temporal resolution. In addition to 6 standard 7-day
196 Lanzoni traps (three regional VPPS2000 aerobiological stations and three VPPS2010 traps in the
197 close vicinity of local source), which had drums rotating at 2 mm h⁻¹, allowing for maximal 1 h
198 temporal resolution, we decided to collect pollen above the source using Burkard Scientific
199 SporeWatch samplers that allowed adjusting the full rotation of the rotating drum to 21 h resulting
200 in maximal 7.5-minute temporal resolution of the samples. Two SporeWatch samplers were
201 installed in the centre of the *A. artemisiifolia* pollen source area. One was set to sample air at 1.5 m
202 above ground level, which is 0.5 m above the canopy of *A. artemisiifolia* plants, and the other was
203 set to sample air at 6 m above ground level. Since the footprint area increases with the height of the
204 sensor and the contribution of the area under the sensor decreases (Burba and Anderson, 2005), the
205 lower SporeWatch sampler was expected to collect pollen from the experimental pollen source area.
206 The SporeWatch sampler installed at 6m above the ground was expected to provide data for
207 exploring the concentration profile in case of notable vertical wind velocities (i.e. turbulence),
208 while, in case of notable horizontal wind velocities, it is expected to give information on
209 background levels of *A. artemisiifolia* airborne pollen concentrations together with Lanzoni traps. In
210 2015, on 1st September and 2nd September, the full rotation of the rotating drum in SporeWatch
211 traps in the middle of the experimental field (both at 1.5 m and 6 m above ground level) was set to 3
212 h, resulting in maximal 1.07-minute temporal resolution of the samples. This allowed us to get

additional insight in the variability of airborne pollen concentrations during the period of high emission (6:00-12:00).

Collected samples from both types of samplers (Burkard and Lanzoni) were prepared and analysed to retrieve average pollen concentrations per unit of time following the European Aerobiology Society's minimum recommendations (Galán et al., 2014) and expressed as pollen grains per cubic metre of air (pollen grains m^{-3}) (Galán et al., 2017). After the removal from the trap, the tape was divided into 48 mm segments. This corresponded to 24-h periods (full rotation of the drum every 168 h, i.e. 7 days), 3-h periods (full rotation of the drum every 21 h) and 25.71-minute periods (full rotation of the drum every 3 h). Each segment was mounted between a glass slide and a cover slip using glycerine-gelatine stained with basic fuchsin. The slides were examined using light microscopy ($\times 200$ magnification) by counting all pollen grains of *A. artemisiifolia* as the slide is scanned along three longitudinal transects ($\approx 20\%$ of the total area). The counts were annotated for every 2 mm segment to allow calculation 1-h, 7.5-minute and 1.07-minute concentration, as determined by the rotation speed of the drum and width of the sampling orifice.

Due to technical issues with the pollen and spore traps (i.e. stop of rotation of the drum) some data are missing from the analysis.

2.3. Meteorological data

With the aim of exploring the relationships between environmental conditions and recorded airborne pollen concentrations, a meteorological station with temperature (T), relative humidity (RH), solar radiation (SR) and precipitation (PP) sensors was set in the middle of the experimental pollen source at 1.5 m above ground level (0.5 m above the canopy) while a 3D sonic anemometer was set at 2.5 m above ground level (1.5 m above the canopy) (Table 1). The location of meteorological sensors in the immediate vicinity of the pollen source was expected to provide the data representative for the field conditions related to anthesis and pollen emission. Therefore, the relationships between measured pollen concentrations and meteorological parameters should reveal conditions that drive *Ambrosia* pollen emission processes. To know them is a prerequisite for determining the function for mechanical description of the emission suitable to be used in dispersion models.

Table 1. List of the equipment used for collecting data and the measurement characteristics.

Sensor	Manufacturer (and type of device)	Pcs.	Temporal resolution	Height (above ground level)	Locality and distance from the pollen source area	Information range
Pollen and spore trap	BURKARD SCIENTIFIC SporeWatch	2	7.5 min ¹	1.5 m	Backi Maglic (BM), center	Local
				6 m	BM, center	Local and Background
	LANZONI VPPS2000	3	60 min	1.5 m	BM, 20 m NW ²	Background
				1.5 m	BM, 15 m E ²	Background
				1.5 m	BM, 60 m E ³	Background
	LANZONI VPPS2010	3	60 min	roof-level	Sombor	Regional
				roof-level	Vrbas	Regional
				roof-level	Novi Sad	Regional
3D sonic anemometer	YOUNG 8100	1	20 Hz (2014) 10 Hz (2015-2016)	2.5 m	BM, center	Local
Thermo-Hygro Sensor + Shelter	YOUNG 41382+41003	1	1 Hz	1.5 m	BM, center	Local
Tipping Bucket Rain Gauge	YOUNG 52203	1	Yes/No rain (2014) 1 Hz (2015-2016)	1.5 m	BM, center	Local
Global Radiation Sensor	WILMERS Messtechnik	1	1 Hz	1.5 m	BM	Local

¹except on 1st and 2nd September 2015 at 6-12 AM when full rotation of drum set to 3h resulting in maximum resolution 1.07 min;

²only in 2015; ³in 2015-2016

Zink et al. (2013) identified temperature, relative humidity, turbulent kinetic energy and precipitation as the key factors for the parameterization of pollen emission processes. In order to examine the influence of the wind on pollen concentration we calculated the turbulent kinetic energy (TKE), as a measure of turbulence that lifts the pollen into the air. TKE was calculated per unit mass (Stull, 1988) as the sum of variances of all three wind components, u , v and w , in x , y and z direction, respectively (Equation 1).

$$TKE = \frac{1}{2} (\overline{(u')^2} + \overline{(v')^2} + \overline{(w')^2}) \quad (\text{Equation 1})$$

During the 2014 campaign, data from the tipping bucket rain gauge were impacted by a technical malfunction (the rain gauge was not unblocked properly at the setup), so the data can only be interpreted in a qualitative manner (yes/no precipitation). Also, the frequency of the sonic measurements was set at 20 Hz which was at the limit of the receiving rate of the data logger (WILMERS Messtechnik GmbH NDL485). Consequently, the actual receiving rate and thus the time resolution were not stable and fluctuated considerably but allowed estimation of dominant wind directions needed for setting Lanzoni VPPS2010 traps in the vicinity of the pollen source in 2015 and 2016. Observed dominance of NW and E wind directions was confirmed by measurements also in 2015 and 2016. Since sonic data were not complete for calculating TKE, the analysis of the relationship between TKE and pollen concentrations was performed on 2015-2016

dataset when wind measurement frequency was set to 10 Hz, which ensured the logger was able to receive the data from Sonic at any time resulting in a constant temporal resolution.

2.4. Data analysis

The study was focused on emissions from a known source, measured at 0.5 m above the canopy (1.5 m above the ground). The analysis was performed using Python analytical libraries (JetBrains, 2017) and Microsoft Excel. Timestamps where data were missing (i.e. pollen concentration or meteorological parameters) were omitted from the analysis. All timestamps used in this paper are presenting local time, in CEST time zone. Meteorological data were averaged within 7.5-minute intervals before the analysis.

At first, stationarity, periodicities and normality of the data were explored (Bendat and Piersol, 2011). Due to the existence of the daily release cycle, the pollen emission process is time variant. This can also be seen in our dataset. To confirm non-stationarity of the pollen dataset we used the non-parametric reverse arrangement test that does not require any prior knowledge about the distribution of the pollen release (Bendat and Piersol, 2011). The presence of periodic components in the signal of pollen emission was detected by a visual inspection of the autocorrelation function which showed evident periodic peaks within 24-hour period and a very slow decrease for different time lags due to the non-stationarity of the data. As identified by the D'Agostino-Pearson test (Pearson et al., 1977), the distribution of pollen concentration signal deviates from the Gaussian distribution. It is interesting to note that even after removing non-stationarity from the data using a composition of Box-Cox transformation (Brockwell and Davis, 2016) and differencing, data still deviated from the normal distribution. Therefore, all analysis was conducted on the raw dataset with non-parametric statistics, while the implication of non-stationarity is discussed in a later section.

Kendall rank correlation coefficient (τ) was calculated as a measure of statistical dependence (Kendall, 1938) between the pollen concentration and meteorological data. The correlation analysis was performed for entire days, on data limited to 6:00-16:00, which is the period when major anthesis are expected (Martin et al., 2010) and on data limited to 9:00-12:00, when the maximum emission is expected. Limiting the analysis to the main emission period aimed at preventing the overestimation of the intensity of correlations resulting from existence of long periods with very low values before the start and after the end of the daily emission (Sikoparija et al., 2018).

298 In order to validate the findings of laboratory experiments by Bianchi et al. (1959) and the
299 emission parameterization designed by Zink et al. (2013), we tested the distribution of 7.5-minute
300 pollen concentrations for the ranges of temperature (in the intervals of 1 °C), relative humidity (in
301 the intervals of 5%) and TKE (in the intervals of 0.1 m² s⁻²). Since the analysis strived to explore
302 the characteristics of the emission from the source, we focused on the period from 6:00 to 16:00 as
303 the main emission period for *Ambrosia* (Martin et al., 2009; 2010).

304 Parameterization of the pollen emission requires a function for each meteorological variable
305 and pollen, as proposed in EMPOL (Zink et al., 2013). Since EMPOL was originally made for the
306 emission of birch pollen, but adaptable for different plant species, its authors highly recommended
307 using field experiments for tuning the parameter values in the functions. Here, we compared the
308 dependencies obtained from our measurements to the functions given in EMPOL. The relationships
309 that were not in accordance to functions given in EMPOL were assessed in detail and
310 parameterized. Curve_fit function from the *scipy.optimize* library in PyCharm (JetBrains, 2017) was
311 used to get the first guess values. Then we fine-tuned the model by varying the function parameters
312 to minimise the mean squared error between the calculated values and the average pollen
313 concentration, normalized to the maximum value from the distribution. Since we focused on the
314 emission from the plants, our parameterization was restricted to the daytime (i.e. solar radiation
315 greater than zero). Nevertheless, it was important to inhibit non-realistic emission in the afternoon
316 hours. This was achieved by introducing a function of diurnal emission cycle, which was calculated
317 as 1-y at each time step, where y is the percentage of the daily integral of airborne pollen
318 concentration (calculated from 7.5-minute measurements) released until a given timestamp.
319 According to common practice, the daily integral of airborne pollen concentration is estimated from
320 the pollen calendar (Sikoparija et al., 2018). Since there is a large uncertainty in the estimated
321 pollen production, the emission-parameterization model was evaluated for the daily emission
322 capacities ranging from minimum to maximum values of pollen production. Daily emission
323 capacity corresponds to the amount of pollen that can be emitted per day under perfect conditions
324 (i.e. $Q_{pollen,day}$ in EMPOL).

325 Finally, the results from the model were compared to measurements from 8 days without
326 precipitation selected from the peak of the pollen season in 2015 and 2016. These data were left out
327 from the model calibration. The first performance criterion was the conservation of mass, for which
328 we compared the daily integral of airborne pollen concentration (X) of modeled and measured data,
329 by calculating the relative error (Equation 2).

330
$$relative\ error = \frac{ABS|X_{modeled}-X_{measured}|}{X_{measured}} \times 100\ (%) \quad (Equation\ 2)$$

The second one was the simultaneous occurrence of airborne pollen which we assessed by performing correlation analysis, using Pearson and Kendall rank correlation coefficients. High correlation indicates more simultaneous variation of the measured and modeled values. The correlation coefficients are sensitive to the start of the emission and the shape of the daily profile.

3. Results

3.1. Average pollen production per plant

The average pollen production per plant in our experimental source was estimated to be 2.55×10^9 pollen grains (minimum = 2.68×10^8 and maximum = 1.59×10^{10}). Considering that 1-3 male flowers per inflorescence mature and release pollen per day, and assuming that this takes place in all male heads of all plants in our source, we have estimated possible daily emission capacity for our source to range from 6.38×10^9 to 7.70×10^{11} pollen grains and used these values for testing the emission parameterization model.

3.2. Influence of the source

Tenfold difference between records at different heights above the source (Fig. 2A and 2B) confirms that the influence of background pollen on records at 0.5 m above canopy can be considered neglectable and thus the airborne pollen recorded at 0.5 m above the source was taken as representative for the emission. The influence of experimental *A. artemisiifolia* pollen source to measurements at 0.5m above the canopy is also supported by tenfold lower records in Lanzoni VPPS2010 traps measuring at 1.5 m in dominant wind directions around the source and by further decrease in concentrations recorded in regional pollen traps situated at roof level in Novi Sad, Vrbas and Sombor (Fig. 2C and 2D).

There was a simultaneous sharp increase of pollen concentrations in both traps situated above the ragweed canopy (i.e. 0.5 m and 5 m), when the source was disturbed by mowing on 2nd September 2015 and 1st September 2016 (Fig. 2 grey bars). This indicated a notable influence of our source on records in both traps situated above the ragweed canopy. The dominance of our artificial source was further confirmed by a sharp decrease in measurements at 0.5 m above the canopy on 3rd September 2015 following the total removal of the source and on 2nd- 5th September 2016 following the 50% reduction of the source.

(A)

(B)

(C)

(D)

Fig. 2. Daily pollen concentrations (pollen grains m^{-3}) (A) measured by Burkard Scientific SporeWatch trap at 0.5m above the canopy, (B) measured by Burkard Scientific SporeWatch trap at 5m above the canopy (C) in the background air represented by average from measurements using Lanzoni VPPS2010 traps at 1.5m above the ground in vicinity of pollen source area and (D) in the regional air represented by average from measurements using Lanzoni VPPS2000 traps at roof level in Novi Sad, Vrbas and Sombor (grey columns in A and B indicate days when source was disturbed by mowing, also note tenfold difference in scale of y-axis).

3.3. Seasonality of the pollen emission and dispersion

There is a clear seasonality in the time series of daily pollen concentration resulting from regularity in *A. artemisiifolia* flowering phenophase (Fig. 2). Because of the elimination of the source in the end of 2015 and 2016 seasons, the best representation of the full flowering season was given in 2014. Pollen emission has a bell shape with the start at the end of July, peak at the end of August and ends in mid-September. Analysed time series show statistically significant positive correlation (Kendall rank correlation coefficients (τ) ≥ 0.62 , p-value $\leq 3.6 \times 10^{-15}$) confirming their representativity for depicting progress of the *A. artemisiifolia* flowering phenophase in the study region.

3.4. Hourly pollen distribution

Similarly, there is a regularity in the diurnal cycle (Fig. 3). Sharp increase in the emission starts at about 7:00 and after the peak is reached at about 9:00-11:00, concentrations gradually decrease. It is interesting to note that pollen was recorded even in the evening hours with distinctive peaks until 23:00. The increase in temporal resolution from 1 h to 7.5 minutes reveals that during the first peak, alternating periods of higher and lower concentrations are recorded. This suggests puffed mode of release rather than continuous release. If the sampling resolution is further increased to 1.07 minutes, additional variability in recorded concentrations can be seen during the main emission period (Supplementary material Fig. S2).

396

397 Fig. 3. Boxplots of pollen concentration (pollen grains m^{-3}) depicting the diurnal cycle of the pollen
 398 emission at 0.5 m above the canopy in the source area at (A) 1 hour and (B) 7.5-minute temporal
 399 resolution (whiskers are set to 5% and 95% while horizontal lines denote the median).

400

401 3.5. Influence of meteorological conditions

402

403 Meteorological parameters such as T, SR, TKE directly correlate with pollen emission while RH
 404 and PP correlate indirectly (Table 2). However, the applicability of correlations when analysing
 405 relationships between pollen data and meteorological conditions, that are expected to influence the
 406 emission, is limited. Long data series result in high significance of correlations ($p < 0.01$) but the
 407 intensity of the correlation is weak (Table 2). Further decrease in intensity when focusing on the
 408 main anthesis period (6:00-16:00) or the maximum emission period (9:00-12:00) suggests that there

409 is a low statistical linear dependence between meteorological conditions and pollen concentrations
 410 at 7.5-minute temporal resolution. Some correlations calculated for the maximum emission period
 411 are even statistically insignificant. Further analysis was therefore focused on understanding the
 412 simultaneous influence of different meteorological variables on pollen concentrations.

413

414 Table 2. Kendall rank correlation coefficients (τ) between meteorological parameters (T, RH, SR,
 415 TKE, PP) and pollen concentrations measured at 7.5-minute resolution and 0.5 m above the canopy
 416 in the source area, for three different periods. All values are significant at the 0.01 level (two tailed)
 417 if not underlined.

418

Dataset focused on 0:00-24:00

419

	Pollen	T	RH	SR	TKE	PP
Pollen	1					
T	0.27	1				
RH	-0.28	-0.74	1			
SR	0.42	0.51	-0.52	1		
TKE	0.31	0.30	-0.30	0.50	1	
PP	-0.03	-0.12	0.14	-0.04	0.07	1

423

Dataset focused on major anthesis period 6:00-16:00

424

	Pollen	T	RH	SR	TKE	PP
Pollen	1					
T	0.10	1				
RH	-0.09	-0.76	1			
SR	0.13	0.60	-0.62	1		
TKE	0.09	0.22	-0.25	0.36	1	
PP	-0.10	-0.16	0.18	-0.17	0.06	1

428

Dataset focused on maximum emission period 9:00-12:00

429

	Pollen	T	RH	SR	TKE	PP
Pollen	1					
T	0.05	1				
RH	<u>-0.02</u>	-0.66	1			
SR	<u>-0.01</u>	0.50	-0.53	1		
TKE	<u>0.02</u>	-0.12	0.06	0.11	1	
PP	-0.09	-0.16	0.17	-0.16	0.09	1

434

435 The distribution of pollen with temperature shows the expected profile, given as the product
 436 of two sigmoid functions, with maximum values in the interval 20-24°C (Fig. 4A). After a detailed
 437 day-by-day analysis, we could not identify limiting temperature thresholds that stop the emission
 438 once the daily cycle started, indicating that the correlation is rather statistical than causal. Both
 439 pollen emission and temperature may be influenced by other factors such as daylight.

440 The distribution of pollen with relative humidity shows that the largest emission is
 441 concurrent with the relative humidity ranging in the interval 55-85% and that the values around

95% can prevent the emission (Fig. 4B). The examples on how high relative humidity delays or even switches off the emission after its start are given in the Supplementary material (Fig. S3).

Despite expecting that the puffed mode of release would significantly depend on the energy available for the emission of pollen from its reservoir, there is no obvious regularity in pollen distribution depending TKE, which in most cases was in the interval 0-0.5 m² s⁻² (Fig. 4C). Although one could expect that the higher energy available for shaking the source and lifting the pollen will result in higher pollen concentration, the relationship is not direct. Therefore, if other factors do not prevent emission, the values of TKE even less than 0.1 m² s⁻² are providing sufficient energy to lift the pollen into the air.

There were only 11 days with precipitation in the study period of years 2015 and 2016. As expected, the rain removes pollen from the atmosphere and from the leaves and other surfaces. However, measurements showed that the start of the rain often resulted in a sharp increase in emission. The rain coincides with instability of the atmosphere that is shown by an increase of TKE. Also, rain drops can physically shake the source and initiate the emission of pollen that remained in the flowers, or lift the pollen that is accumulated on leaves and surrounding surfaces. This is clearly depicted in two consecutive days (10th and 11th August 2016) when it rained in the evening hours following the normal morning emission (Supplementary material Fig. S4). On the first day, arrival of the front caused instability of the atmosphere which resulted in pollen emission in the evening hours at 19:00-20:00. However, notable instability at 7:00 on the second day did not result in the emission because former rain episodes washed out all pollen from the surrounding surfaces and the relative humidity did not decrease under the threshold required for anthesis and emission from ragweed flowers. Later in the evening hours on 11th August, another event of rain with coinciding instability of the atmosphere resulted in the emission of pollen at 17:00-18:00, thus confirming the importance of TKE for the emission of pollen from both the plant and the surrounding surfaces. On the other hand, in Supplementary material Fig. S3A shows an example of increase in pollen concentration when rain started, although the TKE notably decreased.

476
 477 Fig. 4. 7.5-minute pollen average concentration (columns) and its number of records (circles) at
 478 intervals of recorded (A) T and (B) RH for all three seasons and (C) TKE in 2015-2016. The
 479 numbers on the x-axes represent the lower boundary of the class.

480
 481
 482
 483
 484

3.6. Parameterization of TKE function and model results

Field measurements confirmed that the distribution of pollen with temperature and the limiting effect of relative humidity are conceptually in accordance with functions developed for EMPOL emission parameterization (Zink et al., 2013), yet this was not the case regarding the influence of TKE (Fig. 4). Parameterization of the influence of TKE needed modifications since measurements showed that very low values of TKE can initiate the emission, sometimes with very high concentrations of pollen (Fig. 4C). For that reason we calculated the distribution of the average pollen concentration with TKE at the intervals of $0.01 \text{ m}^2 \text{ s}^{-2}$ and focused on the data from early emission period (06:00-10:00) to ensure the recorded pollen concentrations were predominately related to the pollen release from the flowers (Supplementary material Fig. S5A). Results showed it was possible to keep the original concept from EMPOL and use a sigmoid function (Equation 3).

$$f(x) = \frac{1}{(1+e^{-ax+b})} - c \quad (\text{Equation 3})$$

The first guess values of parameters of the sigmoid function ($a = 4.02728603617$, $b = 0.261944471458$ and $c = 0.40648813275$) came from the fitting of the curve (Supplementary material Fig. S5B). Fine-tuning of the parameters resulted in the function (Equation 4) that was used to calculate emission.

$$f_{E,TKE} = \frac{1}{(1+e^{-4TKE+0.31})} - 0.4 \quad (\text{Equation 4})$$

The model was used to estimate the pollen emission for the 8 days shown in Fig. 5 that were not used in establishing the model. When comparing the measured and the modelled emission, the Pearson correlation coefficient varied notably for different estimations of daily emission capacity and for the different days (Fig. 5A). Strong correlations (greater than 0.6), existed in the range of $45-75 \times 10^9$ pollen grains, while further increase of daily emission capacity resulted in the decrease of correlation coefficient. This indicated a high sensitivity of the model to the daily emission capacity. Kendall rank correlation coefficients were less sensitive to changes in emission capacity yielding moderate or strong correlations (greater than 0.5), in the interval of $5-175 \times 10^9$ pollen grains (Fig. 5B). Relative error dropped below 5% for the daily emission capacity larger than 40×10^9 pollen grains (Fig. 5C).

Fig. 5. (A) Pearson and (B) Kendall rank correlation coefficient between modeled and measured pollen concentrations and (C) the relative error between modeled and measured daily integral of airborne pollen concentration, for the different daily estimation of emission capacity, for the eight days from the peak of the season.

526 4. Discussion

527

528 4.1. Characteristics of the *Ambrosia* pollen source

529

530 Although the same methodology was used, the average production of *Ambrosia* plants in the
531 analysed source was twice the estimate given by Fumanal et al. (2007), which is commonly used as
532 a reference for pollen production potential of the common ragweed. The large variability in pollen
533 production between individual plants and populations is related to multiple factors that can affect
534 plant fitness, ranging from genetic to environmental (Fumanal et al., 2007; Rogers et al., 2006;
535 Simard and Benoit, 2011; Simard and Benoit, 2012). With respect to the daily emission capacity,
536 we found that a common ragweed plant can release on average 2.13×10^8 pollen grains which is a
537 magnitude less than, to our knowledge, the only estimate of daily emission capacity available in the
538 literature (Bouillène and Bouillène, 1930). We must note here that our estimate assumes that all
539 racemes and male heads on the plants release pollen from at least 1-3 male flowers which makes
540 this the “worst case scenario”. Although there is a notable variability in estimated pollen production
541 for common ragweed, the daily emission capacity, when combined with the abundance and the
542 distribution of plants, is a valuable input parameter for numerical dispersion models. Nevertheless,
543 we would like to underline here that the daily emission capacity is not the same as the overall level
544 of the pollen flux because the latter is a product of the total amount of pollen that can be released
545 during a day and the atmospheric conditions that drive pollen release.

546

547 4.2. *Ambrosia* airborne pollen around the source

548

549 The amount of airborne ragweed pollen recorded in regional traps during the study period
550 corresponds to long term records on the Pannonian Plain (Sikoparija et al., 2018), making the study
551 seasons representative for the emission in the area considered as the most infected by ragweed in
552 Europe (Smith et al., 2013). Notably, higher concentrations of airborne *Ambrosia* pollen measured
553 at 0.5 m above canopy in the center of the pollen source area, compared to background
554 measurements, confirms the dominance of the local source and ensures that the variability captured
555 by measurements at the immediate vicinity of the canopy reflect the emission. This is in line with
556 the observation from Raynor et al. (1973), which showed that particles in the planetary boundary
557 layer are vertically well mixed after travelling only moderate distances (0.35 km) from their
558 sources. At least tenfold difference between emission levels and concentrations measured at
559 background and regional level agree with the pollen distribution profile determined for *Ambrosia*

(Raynor et al., 1970). The collected data, apart from being useful for exploring the emission, gives an overview of potential exposure amounts in the close vicinity of a common ragweed pollen source. This further emphasizes the problem that observed substantial differences in concentrations at street and roof levels (Rantio-Lehtimäki et al., 1991; Spieksma et al., 2000) can pose to the analysis of the real-life exposure in dose response surveys, making the personalized sampling a more appropriate means of data collection (Werchan et al., 2018). The observed quantity of airborne ragweed pollen in the vicinity of its source highlights the importance of eradicating ragweed on smaller spatial scales to limit the exposure in inhabited areas (Katz and Carey, 2014). In cities, the canyoning effect of houses and buildings (Peel et al., 2014) could limit the dispersion and thus prolong exposure to allergenic pollen at the breathing level.

4.3. Seasonality and daily cycle

The analysis of airborne pollen concentrations has shown notable seasonality and diurnal regularity. Resulting from the timing of ragweed's flowering phenophase, the seasonality of this phenophase is significantly determined by the photoperiod (Deen et al., 1998). Because *Ambrosia* is a quantitative, short-day species, development of its staminate flowers, as well as the start of the pollination, are expected to be stable for a particular latitude (Storkey et al., 2014). This is further supported by observation that locations in similar latitudes show similar changes in characteristics of flowering that are driven by climate change (Ziska et al., 2011). This behaviour is confirmed when comparing the observed timing of pollen season in our study with the season in the common ragweed's native distribution range. About 10° difference in latitude between Backi Maglic (Serbia) and Tulsa (USA) resulted in about 10-day difference in the timing of the pollen season (Howard and Levetin, 2014).

There is a clear diurnal cycle of *Ambrosia* airborne pollen emission. The majority of pollen recorded in the period from 6:00-12:00 is in agreement with previous studies (Martin et al., 2010; Ogden et al., 1969) and corresponds to biological mechanism behind anthesis (Martin et al., 2009). As shown before (Martin et al., 2010; Ogden et al., 1969) the shape of the diurnal concentration signal is notably affected by meteorological conditions. Our study clearly showed that relative humidity is responsible for delaying emission resulting in later record of the modeled peak by Martin et al. (2010). To our knowledge this is the first study that continuously measured airborne pollen concentrations at temporal resolution below 1 h. Our measurements above the source captured the bimodal shape of the pollen concentration signal, which results from the primary emission from anthesis, followed by emission driven by pistilodium growth (Martin et al., 2009). The increase of the temporal resolution from 1 h to 7.5 minutes provides more precise information

about the presence of the second peak and reveals that during the first peak alternating periods of higher and lower concentrations are recorded (Fig. 3B). In addition, there is a concentration peak commonly recorded in the evening hours. It is important to emphasize that recent findings in the UV-based regulation mechanism for plant circadian clock (Meyer et al., 2017) suggest that an internally-driven daily cycle could be considered as a crucial feature of the ragweed pollen emission process and could be used as a feature in model development with an equal weight to other external drivers (i.e. temperature, relative humidity etc.), like in the case of emission parameterization for SILAM (Prank et al., 2013).

When temporal resolution of measurements is further increased from 7.5 minutes to 1.07 minutes the signal reveals notable variability suggesting that the *Ambrosia* field is not a continuous, but rather puffed source that emits in a number of short term periods, when both plant internal and environmental conditions are appropriate. High-frequency turbulence may lead to the puffed source nature of the *Ambrosia* field. Atmospheric mixing is expected to eliminate this variability in roof-level measurements. Further exposure studies will have to take into consideration such short-term variations in concentrations near the source.

4.4. Parameterization of emission

Low intensity of Kendall correlations between pollen concentration and meteorological parameters is expected since it represents the association between two different physical processes with different dynamics of change (e.g. fast changes of pollen concentration between consecutive timestamps are followed by slow changes of temperature, relative humidity etc.). The signs of relationships are in line with the effects that meteorological factors are expected to have on emission, i.e. humidity and precipitation hinders emission and pollen dispersion while temperature and TKE foster the emission and dispersion (Table 2), as it is already shown for ragweed in several aerobiological studies (Bartkova-Scevkova, 2003; Peternel et al., 2006; Puc, 2004). However, as suggested by previous studies (Ritenberga et al., 2016; Sikoparija et al., 2018; Sofiev et al., 2015), the interpretation of the results of correlation analysis between the pollen concentration and meteorological parameters is difficult because the time series of pollen concentrations represents a strongly non-stationary and non-ergodic process or because the relationship between pollen and meteorological conditions might be nonlinear (Jones and Harrison, 2004) and depend on simultaneous effect of several meteorological parameters.

We have confirmed that the functions describing the influence of temperature and relative humidity on pollen emission given in EMPOL (Zink et al., 2013) are in accordance with the

628 distribution of our data (Fig. 4). Still, EMPOL was originally designed for the emission of birch
629 pollen. To adapt the EMPOL emission parameterization to ragweed, we calculated new parameter
630 values for the sigmoid function of emission depending on TKE using our measurements. However,
631 direct implementation of our new function in EMPOL is not possible because our function is based
632 on calibration of the data 0.5 m above the source and EMPOL was designed to be used in numerical
633 dispersion models that typically have their lowest model level at around 10m and horizontal
634 resolutions in the range of km.

635 Furthermore, we introduced a new function, describing the diurnal emission cycle, based on
636 the daily integral of airborne pollen concentration. A similar concept of emission based on diurnal
637 variations in pollen release has been presented by Prank et al. (2013).

638 We tested the estimated emission capacity with the new parameterization. Pearson
639 correlation coefficient seemed to be stricter than Kendall rank correlation coefficient, giving a more
640 suitable, narrow interval of the estimated emission capacity, for the 8 days selected. With the
641 relative error taken into consideration, $45\text{--}75 \times 10^9$ pollen grains per day are deemed as the most
642 reliable interval of emission capacity for the whole field, to be used in the model. This interval is a
643 very small segment from the beginning of the estimated range, which is not surprising since the
644 range was the result of the “worst-case scenario” when all inflorescences in all plants release pollen.
645 However, from Fig. 5A it is noticeable that for different days, maximal Pearson correlation
646 coefficient is obtained for different values of the emission capacity, indicating the variability in
647 pollen emission from day to day, even in the peak of the season. The increased CO₂ levels can have
648 a notable effect on pollen production (Rogers et al., 2006; Ziska and Caulfield, 2000) which makes
649 generalization of emission capacity to different regions difficult without an appropriate
650 experimental dataset.

651 We would like to underline here that in numerical weather prediction models (e.g. COSMO-
652 ART, in which EMPOL is used as emission parameterization scheme) TKE is calculated at around
653 10m above the ground applying sophisticated schemes and representative for a large area (the order
654 of km²). Therefore, using our function for TKE based emission parameterization in such models is
655 not possible. Emission parameterization could be based on other, nonlinear methods (e.g. artificial
656 neural networks).

657

658 5. Conclusions

659

660 Concentrations of airborne pollen at 0.5 m above the canopy (1.5 m above ground level) of a
661 homogenous field of *A. artemisiifolia* of $10 \times 10 \text{ m}^{-2}$ surface are roughly 20 times higher than

regional and background values, and thus are deemed suitable for exploring emission characteristics of the given source. The increase in sampling resolution of the Burkard Scientific SporeWatch revealed a notable amount of variance that remained hidden by the 1 h resolution of the standard Hirst-type volumetric sampling. The pollen concentration signal supports puffed mode of release rather than continuous one. The correlation between 7.5-minute airborne pollen concentrations and meteorological parameters was weak and hence the analysis suggests nonlinear relationships between meteorological conditions and pollen concentrations above the source.

It was important to introduce the function of diurnal emission cycle into the parameterization of pollen emission. This function is based on the daily integral of airborne pollen concentration, which can be estimated as the product of average daily pollen concentration and the number of records, resulting from the sampling resolution during the day. In the areas where long term aerobiological studies are performed, average daily pollen concentration can be taken from pollen calendar.

We developed a function that describes the emission of *Ambrosia* pollen based on TKE. It gives strong correlations with measurements assuming emission capacity of $45\text{--}75 \times 10^9$ pollen grains per day and for the whole field. The strong influence of the emission capacity on the model performance suggests further studies on the variability of this parameter in the future. Global maps of *Ambrosia* biomass, possibly obtained by advanced remote sensing techniques, would be of great interest.

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697

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Supplementary Material

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Graphical Abstract (for review)

Highlights

- Concentrations above the source are roughly 20 times higher than background values
- Pollen production per day was in range from 6.38×10^9 to 7.70×10^{11} pollen grains
- Shift in temporal resolution from 1 h to 7.5 minutes reveals puffed mode of release
- Turbulent kinetic energy less than $0.1 \text{ m}^2 \text{ s}^{-2}$ is sufficient for lifting pollen

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