

## AEROSOL PARTICLE FORMATION IN THE LITHUANIAN HEMI-BOREAL FOREST

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Aerosol particle observations are needed to determine the conditions of particle formation and growth in different environments. This research focuses on new particle formation (NPF) events in the hemi-boreal forest environment at the Aukštaitija Integrated Monitoring Station (IMS) (55°26'N and 26°04'E, 170 m above the sea level). The parameterisation of aerosol particle Nucleation I (3–10 nm), Nucleation II (10–20 nm) and Aitken (20–100 nm) modes was performed, their inputs to the total particle number concentration (PNC) and distribution were assessed. It has been estimated that around 40% of days in spring and 22% in summer were NPF event days. The highest contribution of Nucleation mode I aerosol particles was observed in June, reaching up to 38% of the total PNC values. The mean growth rate (*GR*) and condensation sink (*CS*) values at the Aukštaitija IMS in April and May were 2.9 nm·h<sup>-1</sup>, 1.30·10<sup>-3</sup> s<sup>-1</sup> and 5.3 nm·h<sup>-1</sup>, 1.35·10<sup>-3</sup> s<sup>-1</sup>, respectively. The *GR* and *CS* values were well in agreement with the results obtained from other hemi-boreal forest sites in the Baltic Sea region.

**Keywords:** new particle formation, nucleation, SMPS, hemi-boreal forest

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### 1. Introduction

As our planet year after year is experiencing a record breaking temperature anomaly (see <https://berkeleyearth.org/global-temperatures-2017/>), aerosol studies from various locations are important to understand how fast the environment is changing and how the nature adapts to the climate change. One of the natural processes of aerosol particle number concentration (PNC) increase in the atmosphere is through the nucleation. It can be observed in different environments: on coastlines [1, 2], in forested areas [3–5] and in other vegetation habitats [6, 7]. Terrestrial vegetation significantly contributes to new particle formation (NPF) by releasing into the atmosphere a vast array of precursors, which start the initial process of nucleation: biogenic volatile organic compounds (VOCs), es-

pecially monoterpene emission by trees [8, 9], iodine (I) and dimethyl sulphide ((CH<sub>3</sub>)<sub>2</sub>S) sources from algae activity in the marine and Arctic environments [10, 11]. This process involves the sub-nanometre cluster formation from a condensable vapour, the growth of homogenous clusters to the sizes detectable by aerosol measurement techniques [12, 13]. It is well established that sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) is one of the main chemical compounds in the atmosphere participating in the NPF process [14]. The process of SO<sub>4</sub><sup>2-</sup> ions involvement in the NPF events is fairly well explained in the case of urban environments, where emission sources of SO<sub>2</sub> gases and other atmospheric pollutants are abundant [15–17]. A different situation arises in clean environments, such as hemi-boreal forest, where H<sub>2</sub>SO<sub>4</sub> contribution to NPF and growth is diminished due to the presence of VOCs [9, 18, 19].

Therefore, additional observations are needed to determine the mechanisms by which particles nucleate and grow in different environments.

The aim of this study was NPF observation in the Lithuanian hemi-boreal forest environment, and estimation of the nucleation events during the warm season and the nucleation process characterisation by the aerosol particle formation rate (*FR*), growth rate (*GR*) and condensation sink (*CS*). In addition, the variation in the input of each mode on the total PNC has been estimated. In this study, aerosol particles were subgrouped into Nucleation mode I and Nucleation mode II, in size ranges of 3–10 nm and 10–20 nm, respectively, and Aitken mode (20–100 nm) [20]. It is known that the peak of aerosol particle mode, produced during the nucleation events, resides in the size range below 10 nm [1]. Nucleation mode I mainly represents freshly formed aerosol particles from primary sources, where particle growth is driven through the condensational growth [21]. In the case of Nucleation mode II, the aerosol particle formation can be attributed either to natural or to anthropogenic emission sources [22–25].

## 2. Methodology

### 2.1. Measurement site description

The measurement campaign was performed at the Aukštaitija integrated monitoring station (IMS) (55°27'N and 26°04'E; 170 m above the sea level), located in Aukštaitija National Park, in the North-Eastern part of Lithuania (Fig. 1). The nearest resi-

dential areas are Tauragnai and Utena (total population of 32 000) in the west and Ignalina (population of 6 000) in the southeast. The measurement site was located in the hemi-boreal forest, bordering Utenas Lake in the southwest and Rūgštelėškis Village in the southeast. This hemi-boreal forest is mainly dominated by Scots pines, the rest of the area is filled with Norway spruces and birch trees [26, 27].

### 2.2. Measurement technique

The aerosol particle number size distribution was measured in a size range of 3–110 nm, using a scanning mobility particle sizer (SMPS). It was composed of two units: a condensation particle counter (CPC, TSI model 3776) and a differential mobility analyzer (DMA, TSI model 3085). The SMPS working parameters were the following: aerosol flow  $1.5 \pm 0.1 \text{ L} \cdot \text{min}^{-1}$ , sheath flow  $6.0 \pm 0.1 \text{ L} \cdot \text{min}^{-1}$ , and positive high voltage varied from  $10^1$  to  $\sim 10^4 \text{ V}$ . The duration of the up-scan was 4 min and the whole cycle was repeated every 5 min. A custom impactor (0.071 cm, TSI) placed in front of the SMPS ensured that large particles did not distort the calculations, as an effective cut size diameter ( $D_{50\%}$ ) of 560 nm was well above the measurement size range. Aerosol particles were neutralised by passing through a radioactive  $^{85}\text{Kr}$  neutraliser (TSI model 3077A). The particle diffusion losses were evaluated using the Gormley–Kennedy equation for a cylinder [28]. The multiple charge correction was applied using the AIM software provided by TSI.



Fig. 1. The forest measurement site at the Aukštaitija Integrated Monitoring Station.

A PM<sub>10</sub> inlet was installed 4 m above the ground, attached to a stainless steel tube of 16 mm inner diameter. The total sampled aerosol flow was 5.5±0.1 L·min<sup>-1</sup>. Inside the building this aerosol flow was split into two separate lines. A stainless steel tube of 3 mm inner diameter and a flow rate of 1.5±0.1 L·min<sup>-1</sup> was used by SMPS. However, before the sampled aerosol flow reached SMPS, the aerosol humidity was reduced by using a nafion dryer (Perma Pure LLC, model MD-110-72S-4). The measured data quality control was performed periodically during the measurement campaign. It consisted of aerosol and sheath flow rates verification and CPC zero checking. The DMA maintenance was done once per year using 70 and 150 nm polystyrene particles. An automatic weather monitoring station (DL2e, Delta-T Device Co.) was assembled in the sampling area to measure meteorological parameters locally, such as air temperature and relative humidity (*T* and *RH*; RHT2nl-02 sensor, Delta-T Device Co.). The data were logged at 1-h intervals.

### 2.3. NPF event classification

The classification of the NPF was based on the principles described in Refs. [29, 30]:

**Type I:** The growth of newly formed particles is clearly distinguishable in the particle number size distribution plots. The main physical parameters – growth rate (*GR*) and formation rate (*FR*) – characterising the event can be determined without doubt.

**Type II:** The cluster of new particles is present, however, the shape of the growing particle mode is unclear, and thus the retrieving physical parameters (*GR* and *FR*) are possible only for some of these events.

**Non-event:** The days when no new particles in the Nucleation particle mode were formed.

**Undefined:** It is unclear whether the new particle formation occurred, the event did not fulfil the requirements of the criteria mentioned above.

### 2.4. Physical parameterisation of new particle growth and formation

For each nucleation event, the *GR* was calculated graphically from the contour plots. The *GR* can be expressed as in Ref. [31]:

$$\frac{dr}{dt} = \frac{m_v \beta_m DC}{r\rho}. \quad (1)$$

Here *r* is the particle radius, *m<sub>v</sub>* is the molecular mass of condensable vapour, *β<sub>m</sub>* is the transitional correction factor for mass flux, *D* is the diffusion coefficient, *C* is the concentration of the condensable vapour, and *ρ* is the particle density. Eq. (1) can be integrated from *r<sub>0</sub>* to *r* to obtain the condensable vapour concentration:

$$C = \rho \left\{ \frac{r^2 - r_0^2}{2} + [4/(3\alpha) - 0.623]\lambda(r - r_0) + 0.623\lambda^2 \ln \frac{\lambda + r}{\lambda + r_0} \right\}. \quad (2)$$

*α* is the mass accommodation coefficient (i.e. sticking probability) and *λ* is the mean free path. The parameters *dr/dt* and *dN<sub>3nm</sub>/dt* can be obtained directly from the measurements. The condensation sink (*CS*) is *4π·D·CS'*, and *CS'* is integrated over the aerosol size distribution:

$$CS' = \int r\beta_m(r)n(r)dr = \sum_i \beta_m r_i N_i. \quad (3)$$

The *β<sub>m</sub>* is expressed by terms of the Knudsen number (*Kn*) and the sticking coefficient (*α*) as follows:

$$\beta_m = \frac{Kn + 1}{0.377 \cdot Kn + 1 + 4/3 \cdot \alpha^{-1} \cdot Kn^2 + 4/3 \cdot \alpha^{-1} \cdot Kn}. \quad (4)$$

The *Kn* and *α* are typically assumed to be unity. In the molecular regime where *Kn* ≫ 1, we have *CS* ∝ *r*<sup>2</sup>. On the other hand, in the continuum regime where *Kn* ≪ 1, we get *β<sub>m</sub>* = 1 and *CS* ∝ *r*. However, typically tropospheric aerosol also includes the transitional regime and *CS* ∝ *a*, where 1 < *a* < 2. It was equal to 1 for this case.

## 3. Results and discussion

### 3.1. Occurrence of new particle formation events at the Aukštaitija IMS

The measurements of the particle number size distribution in a range of 3–100 nm were performed at the Aukštaitija IMS hemi-boreal forest from 8 April to 7 October 2016. Three distinct particle modes were classified as Nucleation mode I (3–10 nm),

Nucleation mode II (10–20 nm) and Aitken mode (20–100 nm). By applying the NPF classification scheme, the days were grouped into event and non-event days (Table 1). The total number of analysed days was 158. Herewith, in total 36 NPF event days were observed and classified into Type I (12 events) and Type II (24 events). There were 25 non-event and 97 undefined days. The NPF events mostly occurred during the period of April–June 2016. It was estimated that 40% of events occurred during the spring, 22% during the summer and no events were depicted in the autumn due to scarce measurement data. The contribution of each aerosol particle mode to the total PNC was determined (Fig. 2).

Table 1. The classification of NPF event days at the Aukštaitija IMS.

Month	Days				Total
	Type I	Type II	Non-event	Undefined	
APR	7	2	7	7	23
MAY	3	7	7	7	24
JUN	2	9	1	16	28
JUL	0	2	3	20	25
AUG	0	4	4	16	24
OCT*	0	0	3	4	7
Whole season	12	24	25	97	158

\* In October 7 days out of the whole month were covered.

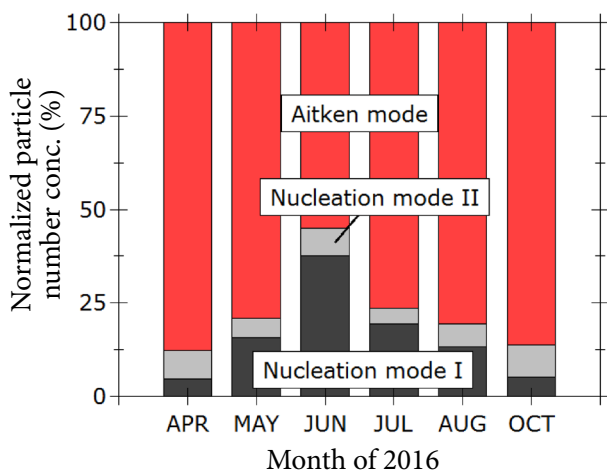


Fig. 2. Aerosol particle mode contribution to the total PNC.

The highest contribution to the total PNC was attributed to the aerosol particles of Aitken mode. It contributed from 55% in June to 88% in April. The contribution of Nucleation mode I varied from 5% in October to 38% in June of the total PNC. Nucleation mode II had no meaningful contribution during the whole measurement campaign. Its contribution varied from 4 to 9% and did not have any clear trend.

### 3.2. Particle number concentration analysis

The total PNC ranged from  $150 \text{ cm}^{-3}$  (5th percentile) to  $14\,960 \text{ cm}^{-3}$  (95th percentile) during the whole measurement period (Fig. 3(a)). The monthly averages of total PNC were lowest ( $235 \text{ cm}^{-3}$ ) in October and highest ( $3\,970 \text{ cm}^{-3}$ ) in August. It should be noted that the fluctuations in the total PNC were also highest during the summer months and lowest in the autumn. The monthly median values were lower than the averages of total PNC in all cases, except for October, thus suggesting that the aerosol particle size distribution is positively skewed. The Nucleation mode I data revealed that the highest variability in particle concentration was observed during the spring season (Fig. 3(b)). During this period most NPF events, classified as Type I (Table 1), had occurred.

One of the highest monthly PNC values in Nucleation mode I were measured in the period from May to July. During that period the 95th percentile lowered from  $3\,620$  to  $2\,160 \text{ cm}^{-3}$  and the highest monthly average ( $1\,150 \text{ cm}^{-3}$ ) was observed in June. Although the mean and median values of Nucleation mode I were higher in the summer than in the spring, the number of NPF events was smaller in comparison to the spring, 40 and 22%, respectively (Table 1). In clean environment the high mean PNC in Nucleation mode I is likely to indicate the presence of a new particle emission source located in the surrounding area. In general, the GR for aerosol particles of sub-10 nm size in the atmosphere is in a range from  $1$  to  $20 \text{ nm}\cdot\text{h}^{-1}$  [31], in this work it was estimated to be  $\sim 5 \text{ nm}\cdot\text{h}^{-1}$ . The transition of aerosol particles from Nucleation mode I to mode II occurred in a 1–2-h period. During the warm period, the air masses are usually transported at speeds from  $20$  to  $60 \text{ km}\cdot\text{h}^{-1}$  in this geographic region [32]. During the transport of air masses from the nearest urban areas, such as Utena and Ignalina, aerosol

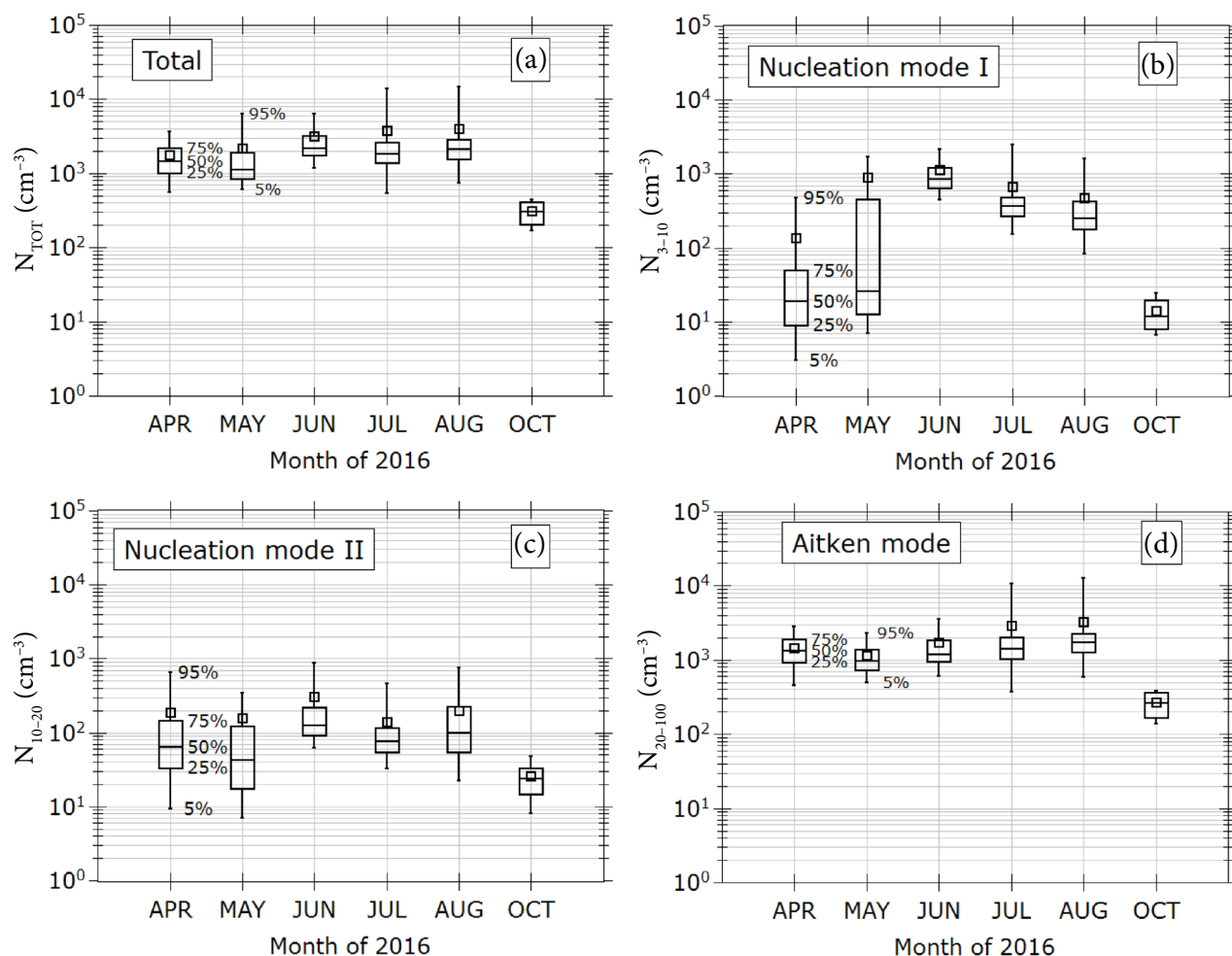


Fig. 3. Box plots of the monthly average PNC of total (a), Nucleation mode I (b), Nucleation mode II (c) and Aitken mode (d). The 5th and the 95th percentiles (whiskers), quartile  $Q_1$  and  $Q_3$  (box), median (horizontal line) and mean (square).

particles would grow substantially to be detected in Nucleation mode II or Aitken mode. Thus, the probable source for increased monthly PNC values in Nucleation mode I during the summer was biogenic activity in the forested area around the Aukštaitija IMS. However, sporadic contamination from local biomass burning sources could not be avoided during the measurement period.

The contribution of Nucleation mode II in the hemi-boreal forest environment was low – the 95th percentile was well below 900 cm<sup>-3</sup> during the whole measurement campaign (Fig. 3(c)). The Aitken mode PNC had the highest value of 3 300 cm<sup>-3</sup> in August (Fig. 3(d)). The Aitken mode data from the Aukštaitija IMS were compared to the aerosol PNC in a size range of 30–100 nm from the stations located in the Northern and Central Europe (Table 2). The stations were

selected according to their type, which is similar to the Aukštaitija IMS designation [33]. The Aitken mode mean PNC (1 800 cm<sup>-3</sup>) during the whole period was one of the highest (except for Preila) in the Northern Europe. For comparison in Central Europe the highest PNC values in this size range were measured at the K-Pusztas and Kosetice stations, 2 700 and 2 420 cm<sup>-3</sup>, respectively. It should be considered that in clean environments the PNC concentrations usually decrease during the autumn [29, 34], and in the Aukštaitija IMS dataset the autumn data was underrepresented. Collecting more data from the SMPS measurement in autumn would have decreased PNC values for the whole campaign, in general. There was a tendency of PNC increase in the Aitken mode during the summer, the same tendency was observed at the forest stations, SMEAR II and Pallas, both

Table 2. Data obtained from Ref. [32], representing the mean values of aerosol PNC in a size range of 30–100 nm for the period of 2008–2009 from the stations located in Northern and Central Europe. The stations are classified by their type and region, the mean, median, 5th and 95th percentiles of 2008–2009 dataset are presented.

Station	Station code	Station type	Region	5th percentile	Mean	Median	95th percentile
Aspvreten	ASP	regional background/forest	Northern Europe	220	1 090	940	2 490
Birkenes	BIR	rural/forest	Northern Europe	60	620	450	1 650
Pallas	PAL	remote/above tree line	Northern Europe	30	370	180	1 310
Preila	PLA	regional background/marine	Northern Europe	350	2 590	1 830	4 930
Aukštaitija IMS *	–	regional background/forest	Northern Europe	440	1 800	1 150	5 470
SMEAR II	SMR	regional background/forest	Northern Europe	150	950	760	2 390
Vavihill	VHL	regional background/marine	Northern Europe	320	1 460	1 280	3 240
K-Puszt	KPO	regional background/rural	Central Europe	800	2 700	2 160	6 390
Kosetice	OBK	regional background/rural	Central Europe	750	2 420	2 030	5 400
Hohenpeisenberg	HBH	alpine/rural	Central Europe	340	1 130	990	2 400

\* In the case of Aukštaitija IMS, the data from the Aitken mode (20–100 nm) during the measurement campaign were compared.

located in Finland [33]. Oppositely, on the Baltic sea coastal environments, the variation in the monthly average PNC of Aitken mode had a general tendency towards increasing in the spring at Preila (Lithuania) [2], Aspvreten and Vavihill (Sweden) [33].

### 3.3. Diurnal patterns of nucleation events

The classical NPF event of Type I was observed on 12 April 2016 (Fig. 4(a)). Fresh particles started to form at 10:30 (GMT+2), followed by a significant growth up to 40 nm in size at 18:00 (GMT+2). Such intense type of particle growth is referred as ‘banana shaped’ particle number size distribution [30]. In April particle burst in the Nucleation modes (Fig. 5(a, b)) usually started at 11:00 (GMT+2) and lasted till the evening, with the peak concentration values reached at 14:00–15:00 (GMT+2). Usually, Type I events start to form around the noon, when the solar radiation

is most intense. Therefore, we refer to these events as midday. Due to the condensation growth and coagulation of newly nucleated particles, the PNC of Nucleation mode II started to increase with 1-h delay, at 12:00 (GMT+2), and reached its highest value at 15:00 (GMT+2). In April the air temperature had the classical trend with reaching the minimum +2.5°C at sunrise and the maximum +11°C at noon. In May the evening bursts of newly nucleated particles were common (Fig. 5(c, d)). Though the initial start of NPF events was still present during the noon, the PNC values in the evening were dominant. In this case PNCs of both Nucleation modes reached maximum values at 18:00 (GMT+2) without a delay. In May the most common events were classified as Type II and due to the time of their occurrence we refer to them as the evening events (Fig. 4(b)). The PNC of Aitken mode was stable and did not vary during the day both in April and in May (Fig. 5(a, c)). In addition, ‘break type’ events, classified as Type II, were

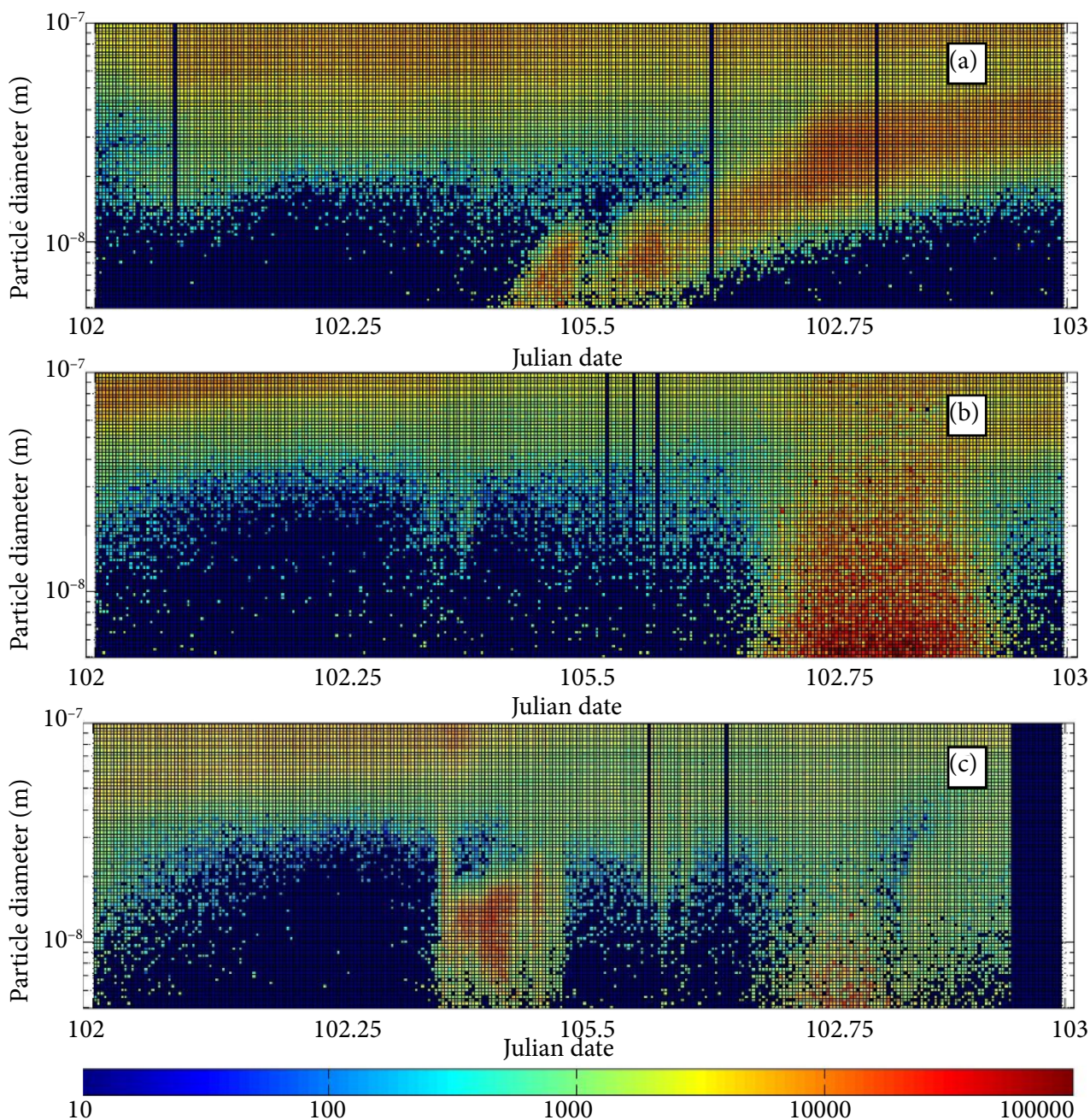


Fig. 4. Examples of NPF events. The midday event, classified as Type I, occurring on 12 April 2016 (a), the evening event, classified as Type II, occurring on 23 May 2016 (b) and the 'breaking type' event, classified as Type II, occurring on 7 May 2016 (c).

observed in May as well (Fig. 4(c)). The 'break type' event was similar to the midday one, however the growth of Nucleation mode particles was not observed in this case. This was due to the fact that the initial NPF event had started not at the measurement site. Newly nucleated particles travelled from the surrounding locations in the vicinity. After the change of the air mass direction, the observation of the nucleation process was terminated.

### 3.4. Parameterisation of aerosol particles

The estimated average growth rate and CS values (Eqs. 1, 3) at the Aukštaitija IMS in April and May were  $2.9 \text{ nm}\cdot\text{h}^{-1}$ ,  $1.30\cdot 10^{-3} \text{ s}^{-1}$  and  $5.3 \text{ nm}\cdot\text{h}^{-1}$ ,  $1.35\cdot 10^{-3} \text{ s}^{-1}$ , respectively. The GR and CS were compared with the results from other measurement sites, located in the hemi-boreal forests of the Baltic Sea region. The average GR and CS values were  $3.9 \text{ nm}\cdot\text{h}^{-1}$ ,  $4.8\cdot 10^{-3} \text{ s}^{-1}$  in the Aspveten (Sweden),

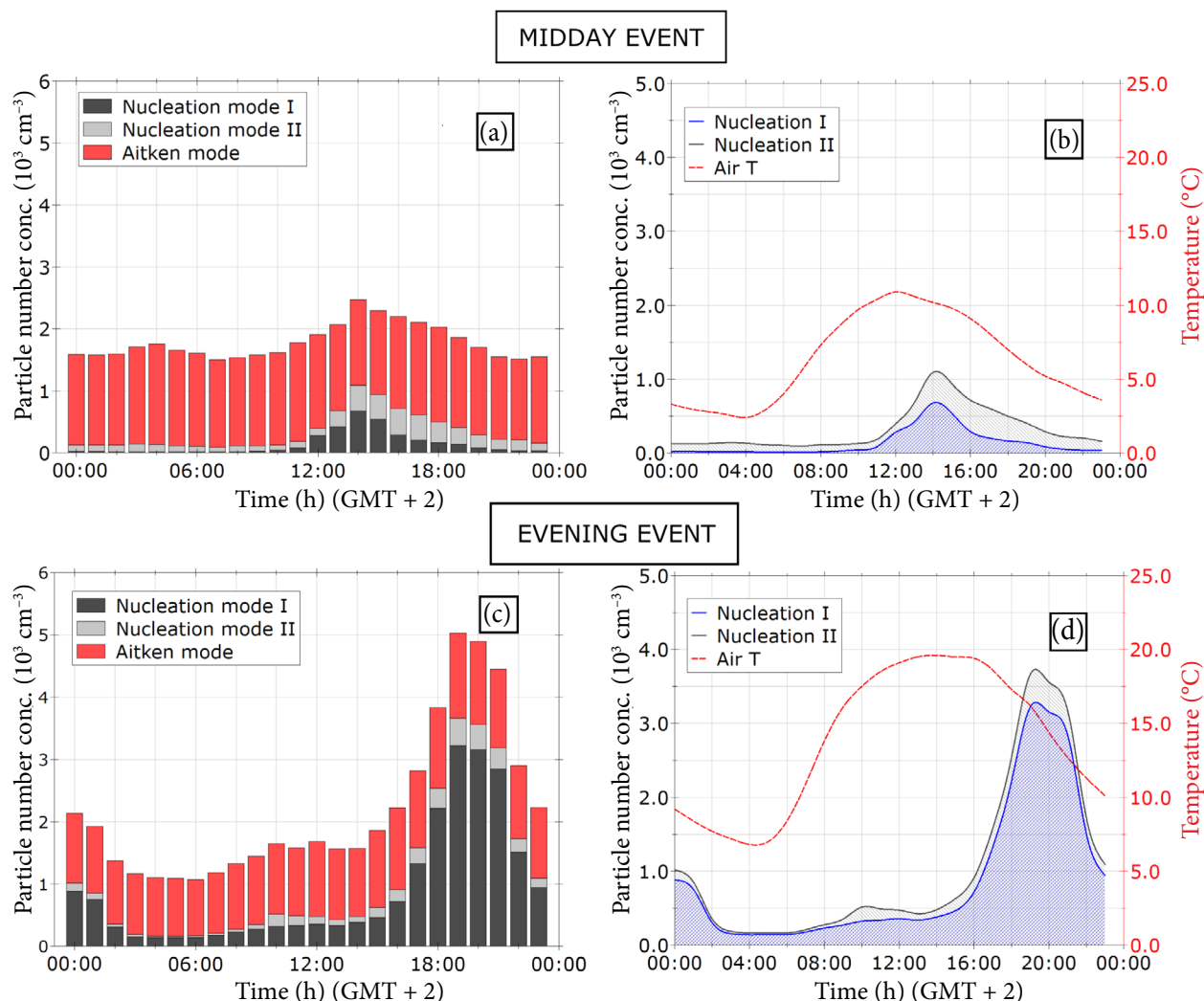


Fig. 5. The diurnal variation of PNC monthly values in Nucleation and Aitken modes during April (a, b) and May 2016 (c, d), the monthly average of air temperature diurnal variation (b, d).

$3.0 \text{ nm}\cdot\text{h}^{-1}$ ,  $3.2\cdot 10^{-3} \text{ s}^{-1}$  in Hyytiälä (Finland) and  $2.5 \text{ nm}\cdot\text{h}^{-1}$ ,  $1.2\cdot 10^{-3} \text{ s}^{-1}$  in Pallas (Finland) stations [4, 30]. In comparison, the *GR* values for the stations situated in the south of the Baltic Sea were higher than the ones from the north. The *CS* mean values from the Aukštaitija IMS were similar to Pallas' ones. This suggests that biogenic VOCs were some of the main factors, influencing the aerosol formation process in this area.

#### 4. Conclusions

During the warm season (April–October 2016), the measurements of aerosol particle number size distribution were performed at the Aukštaitija IMS. The monthly variations in the PNC of Nucleation mode I, Nucleation mode II and Aitken

mode have been studied. The increase of particle number concentration (PNC) values in Nucleation mode I during the summer was caused mainly by biogenic activity in the forested area surrounding the Aukštaitija IMS.

During the investigation period, 36 days (23%) were identified as NPF events: 12 days of Type I and 24 days of Type II. Most of the NPF events occurred in the spring (40% of all days), while in the summer 22% of all days were NPF events.

The contribution of Nucleation mode I (3–10 nm) was always dominating over Nucleation mode II (10–20 nm) during the NPF events. The highest contribution of Nucleation mode I was 38% of the total PNC and was observed in June. Aerosol particles in Aitken mode contributed to the total PNC from 55% in June to 88%



in April. No diurnal pattern for the Aitken mode particles was observed during the investigation period.

The mean aerosol particle growth rate ( $4.1 \text{ nm}\cdot\text{h}^{-1}$ ) and condensation sink ( $1.32\cdot 10^{-3}\cdot\text{s}^{-1}$ ) were well in agreement with the values observed from other forested areas in the Baltic Sea region.

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## AEROZOLIO DALELIŲ SUSIDARYMAS LIETUVOS PUSIAU BOREALINIULOSE MIŠKUOSE

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### Santrauka

Aerolio dalelių susidarymo ir augimo procesų įvairiose aplinkose stebėjimai yra būtini siekiant iširti sąlygas, prie kurių šie reiškiniai vyksta. Darbe nagrinėjamas naujų aerolio dalelių susidarymas Aukštaitijos kompleksinio monitoringo stoties (IMS) miškuose. Įvertinti pagrindiniai fizikiniai aerolio dalelių 1-osios nukleacinės (3–10 nm), 2-osios nukleacinės (10–20 nm) ir Aitkeno modų (20–100 nm) parametrai, nustatytas šių modų bendras indėlis į skaitinį aerolio dalelių dydžių pasiskirstymą ir koncentraciją. Pavasarį buvo nustatyti

40 % naujų dalelių susidarymo atvejai, vasarą – 22 %. Didžiausias 1-osios nukleacinės modos indėlis į skaitinę dalelių koncentraciją siekė iki 38 % nuo bendros koncentracijos, jis buvo stebimas 2016 m. birželio mėnesį. Vidutinės aerolio dalelių augimo greičio (*GR*) ir kondensacinio nuotėkio (*CS*) vertės 2016 m. balandžio–gegužės mėn. sudarė atitinkamai  $2,9 \text{ nm}\cdot\text{h}^{-1}$ ,  $1,30\cdot 10^{-3} \text{ s}^{-1}$  ir  $5,3 \text{ nm}\cdot\text{h}^{-1}$ ,  $1,35\cdot 10^{-3} \text{ s}^{-1}$ . Šio tyrimo metu nustatytos *GR* ir *CS* vertės yra artimos vertėms, gautoms iš kitų Baltijos jūros regiono miškingų vietovių.