An evaluation of new particle formation events in Helsinki during a Baltic Sea cyanobacterial

2 summer bloom

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- 31 Abstract
- 32 Several studies have investigated New Particle Formation (NPF) events from various sites ranging
- from pristine locations, including forest sites to urban areas. However, there is still a dearth of studies
- 34 investigating NPF processes and subsequent aerosol growth in coastal yet semi-urban sites, where the
- 35 tropospheric layer is a concoction of biogenic and anthropogenic gases and particles. The
- 36 investigation of factors leading to NPF becomes extremely complex due to the highly dynamic
- 37 meteorological conditions at the coastline especially when combined with both continental and
- 38 oceanic weather conditions. Herein, we engage a comprehensive study of particle number size
- 39 distributions and aerosol-forming precursor vapors at the coastal semi-urban site in Helsinki, Finland.
- 40 The measurement period, 25 June 2019–18 August 2019, was timed with the recurring cyanobacterial
- 41 summer bloom in the Baltic Sea region and coastal regions of Finland. Our study recorded several
- 42 regional/local NPF and aerosol burst events during this period. Although the overall anthropogenic
- 43 influence on sulfuric acid (SA) concentrations was low during the measurement period, we observed

that the regional or local NPF events, characterized by SA concentrations in the order of 10⁷ 44 45 molecules per cm⁻³ occurred mostly when the air mass travelled over the land areas. Interestingly, 46 when the air mass travelled over the Baltic Sea, an area enriched with Algae and cyanobacterial 47 blooms, high iodic acid (IA) concentration coincided with an aerosol burst or a spike event at the 48 measurement site. Further, SA-rich bursts were seen when the air mass travelled over the Gulf of 49 Bothnia, enriched with cyanobacterial blooms. The two most important factors affecting aerosol 50 precursor vapor concentrations, and thus the aerosol formation, were speculated to be (1) the type of 51 phytoplankton species and intensity of bloom present in the coastal regions of Finland/ Baltic Sea and 52 (2) the wind direction. During the events, most of the growth of sub-3 nm particles was probably due to SA, rather than IA or MSA, however much of the particle growth remained unexplained indicative 53 54 of the strong role of organics in the growth of particles, especially in the 3–7 nm particle size range. 55 Further studies are needed to explore the role of organics in NPF events and the potential influence 56 of cyanobacterial blooms in coastal locations.

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Keywords: coastal environment, particle growth, methane sulfonic acid, cyanobacterial summer bloom, sulfuric acid, iodic acid

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1 Introduction

62 New particle formation (NPF) and growth of aerosols are regional processes occurring globally 63 introducing a substantial aerosol load into the atmosphere. NPF has been observed in different 64 environments, including pristine (Asmi et al., 2016; Jang et al., 2019; Jokinen et al., 2018), polluted boundary layers and urban areas (Kulmala et al., 2021; Kulmala et al., 2017; Manninen et al., 2010; 65 66 Kulmala et al., 2016; Wang et al., 2017; Cai and Jiang, 2017; Deng et al., 2020; Yao et al., 2018; Du 67 et al., 2021; Yan et al., 2021), boreal forests (Buenrostro Mazon et al., 2016; Dada et al., 2017; 68 Kulmala et al., 2013; Kyrö et al., 2014; Leino et al., 2016; Nieminen et al., 2014; Rose et al., 2018), 69 tropical forests (Artaxo et al., 2013; Wimmer et al., 2018) and mountain tops (Bianchi et al., 2016, 70 2020). Few studies have investigated NPF processes in a coastal environment although the coastal 71 NPF research started quite early. The investigation of coastal aerosol events dates back to 1978, when 72 the measurements of total aerosol number concentration were carried out at the Tasmanian coast 73 (Bigg and Turvey, 1978). After that atmospheric nucleation was observed in the Southern hemisphere 74 around the Antarctic coastline (O'Dowd et al., 1997), in Mace Head (Flanagan et al., 2005; McFiggans 75 et al., 2004; O'Dowd et al., 2002), in coastal regions of China and Spain (Yu et al., 2019; Mc Figgans 76 et al., 2010; Mahajan et al., 2011) and in open water regions of North East Greenland (Dall'Osto et al., 2018). Most of these studies have identified biogenic emissions from marine algae as the main precursors driving the new particle formation in a perfect coastal setting.

The measurements of gaseous precursors, meteorology and biogenic influences are important to study the coastal NPF, which may lead to the formation of coastal/marine clouds. Coastal clouds are the drivers of many coastal ecosystem (Carbone et al., 2013, Emery et al., 2018, Lawson et al., 2018). Any impact or fluctuations in the cloud formation may impact several other processes of the fragile coastal ecosystem. These coastal clouds demonstrate a high sensitivity to CCN (He et al., 2021) and they have a significant impact on the radiation budget because they have a high infrared emission and albedo when compared to the dark water bodies down below. In this study we highlight the type of NPF processes and their drivers in a semi-urban-coastal setting where the atmosphere could be a mixture of anthropogenic and biogenic emissions. Unlike the above mentioned previous studies which were mostly carried out in a perfect coastal environments where NPF would be most likely affected by the biogenic emissions, this study helps to evaluate the impact of urban emissions Vs coastal emissions on NPF and at large the cloud formation processes.

It is well documented that sulfuric acid (henceforth SA) is an important precursor to NPF in most environments (Almeida et al., 2013; Kulmala et al., 2013; Croft et al., 2016; Jokinen et al., 2017; Kirkby et al., 2011; Sipilä et al., 2010). The advancement in aerosol research, revealed that a binary system of SA and water is not sufficient to produce particles in ambient atmospheric conditions without stabilizing compounds (Benson et al., 2008; Duplissy et al., 2016; Kirkby et al., 2011). More recently, it has been found that a ternary system involving SA-ammonia-water or SA-amines-water yield much higher nucleation rates as compared to the binary system (Kulmala et al., 2000; Benson et al., 2008; Almeida et al., 2013; Glasoe et al., 2015; Kürten et al., 2016). In addition to these systems, organic compounds which are highly oxygenated - thus less volatile- have been found to contribute to secondary organic aerosol (SOA) mass in forested areas, mountain tops and anthropogenically influenced field sites (Ehn et al., 2014; Pierce et al., 2011; Riipinen et al., 2012; Zhang et al., 2009; Heikkinen et al. 2020; et al., 2020) and laboratory experiments have shown that they can contribute also to the first steps of NPF (Simon et al., 2020; Lehtipalo et al., 2018; Kirkby et al., 2016; Tröstl et al., 2016).

Furthermore, another important molecular class, iodine as well as its related oxidized species play a crucial role in NPF especially in coastal areas (Allan et al., 2015; Mahajan et al., 2009; Raso et al., 2017; Sipilä et al., 2016) and in pristine marine locations (Baccarini et al., 2020; Beck et al., 2021; He et al., 2021). Some previous studies have reported the emissions of I₂ from the macroalgae at coastal sites (Huang et al., 2010; Peters et al., 2005; Saiz-Lopez and Plane, 2004).

Several studies from coastal sites like Roscoff, France (Mahajan et al., 2009; McFiggans et al., 2010), Mace Head, Ireland (O'Dowd et al., 2002) and other European coastlines (Mahajan et al., 2011; Saiz-Lopez et al., 2012) have reported iodine species initiating NPF. The reported events can be considered as aerosol burst events with high aerosol concentration and having exceptionally high initial growth rates (GR) (O'Dowd et al., 2002; McFiggans et al., 2004; Mahajan, et al., 2011). The study from the Roscoff coast suggests that the daytime emissions of I₂ produced by macroalgae during low tides drives the particle formation (McFiggans et al., 2010). The iodine oxides and/or oxoacids formed by the biogenic emissions from the micro- and macroalgae near the coastal regions are capable of selfclustering, which could form new particles with a diameter <3 nm and sometimes with a high gas concentration reaching up to 10⁶ cm⁻³ or even more. Recent studies have shown that ion-induced IA nucleation proceeds at the kinetic limit and the overall nucleation rates (ion-induced nucleation + neutral nucleation) driven by iodine oxoacids (IA and iodous acid, HIO₂) are high, even exceeding the rates of well-known precursors of NPF (He et al., 2021b, 2021a): SA with roughly 100 pptv ammonia under similar conditions (Sipilä et al., 2010). The rapid photolysis of I₂, (< 10 s), produces I atoms above the ocean surface and can be detected in high concentrations close to the source region (McFiggans et al., 2010). However, the compounds with longer lifetimes such as CH₃I (two days) provide a source of iodine throughout the troposphere (Saiz-Lopez et al. 2012).

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Dimethyl sulfide (DMS) oxidation by OH radical in the daytime and by nitrate radical in the nighttime yields other aerosol precursor gases, such as methane sulfonic acid (henceforth, MSA) and SA (Barnes et al., 2006), which play a crucial role in the NPF processes. In a marine coastal environment, MSA concentrations, which are typically lower than those of SA, could be as low as 10% of SA concentration and could maximally reach 100% of SA concentration (Eisele and Tanner, 1993), yet MSA is a potential candidate to participate in the atmospheric nucleation and growth processes (Beck et al., 2021). The stability of heterogeneous MSA clusters have been studied in laboratory and modelling studies (Chen et al., 2020, 2015, 2016) but no study has yet documented MSA clusters in the field. The limited NPF studies in the semi-urban coastal regions and the dynamic coastal meteorology drives the motivation of this research. Another motivation for this research is that, till date no detailed studies on the impact of biogenic emissions on NPF events in Finland were done before in despite the fact that extensive cyanobacteria blooms occur every year in the Baltic Sea region and neighboring water bodies (including Finnish lakes) (Kahru and Elmgren 2014), which could be a significant source of iodine species, SA and MSA. Increasing temperatures and the excessive nutrient load in the Baltic Sea promote algal growth (Kuosa et al., 2017; Suikkanen et al., 2007, 2013). According to HELCOM (Baltic Marine Environment Protection Commission), the Baltic Sea has warmed 0.3° C per decade, however after 1990 significantly faster at 0.6° C per decade and in Finnish coastal areas the warming is even faster with a 2° C increase since 1990 (Humborg et al. 2019). The amount of blue-green algae (i.e. cyanobacteria) has shown a statistically significant increase in open sea areas in the Gulf of Finland, Sea of Åland and the Sea of Bothnia in the last 40 years (Kahru and Elmgren, 2014). The increase in frequency and intensity of cyanobacterial blooms would increase the potential emission of biogenic gases changing the composition of the overlying atmosphere and the atmosphere of the neighboring sites, depending on the meteorological conditions. In this semi-urban coastal setting the concentration of gaseous precursors and aerosol size distribution may be influenced by the local meteorological parameters such as wind direction, wind speed, (air mass) turbulences especially at the surface layer of the lower atmosphere. Coastal locations are dynamic environments with rapid changes in meteorological parameters, also making the study of NPF more challenging.

In this study, we aim at a thorough evaluation of aerosol precursor molecules with a detailed (NPF events) analysis during the cyanobacterial bloom period, in the coastal-city of Helsinki, Finland, from June to August (summer) 2019..This work evaluates the role of phytoplankton blooms and meteorological parameters in the NPF events observed during the measurement period. We also identify the major precursor vapor(s) and molecular clusters found during the aerosol events. Here, we formulate the hypothesis that gaseous precursors formed from the biogenic emissions from the surrounding marine areas could play an important role in the nucleation processes in Helsinki. Although Helsinki is a coastal area yet the role of marine emissions on NPF processes has not been studied before.

2 Measurement Site and Methodology

The measurement sites are surrounded by coastal water bodies (<4km, Vanhankaupunginselkä), forests (<3km) and road connecting to the main city (<300m) as seen in figure 1. Overall Helsinki is located on a relatively flat land on the coast of the Gulf of Finland. The Helsinki Metropolitan area is about 765 km² with approximately one million inhabitants, counting together the city of Helsinki and the neighboring cities of Espoo, Vantaa, and Kauniainen. The climate in southern Finland can be classified as either marine or continental depending on the air-flows and pressure systems. Either way, the weather is milder than typically at the same latitude (60°N) mainly due to the Atlantic Ocean and the warm Gulf Stream.



Figure 1: Map showing the two locations included in the study where instruments were operated (upper left panel). The yellow polygons on the left side of the measurement locations (on the lower right panel) shows forest/park with little or no traffic (West and Northwest, 300 m from the measurement site). The yellow double lines on the right of the measurement locations is the traffic area or the main road (E75) leading to the Helsinki city center (250 m east of the measurement site). The blue lines depict the coastline after which the lakes and coastal waters of Gulf of Finland start (1

The blue lines depict the coastline after which the lakes and coastal waters of Gulf of Finland start (km to the east from the measurement site) © Google Earth 2019

The site and measurement period (25 June 2019–18 August 2019) selected for this particular study are unique since this semi-urban location could be influenced by emissions from the recurring summertime blooms in the Baltic sea and the neighboring coastal regions. As per the SYKE press release (2019) the northern part of the Baltic Sea's main basin, entrance to the Gulf of Finland and south of the Åland Islands, were enriched with blue-green algae (cyanobacteria). The bloom lasted from June-August 2019. In coastal areas, bloom was mostly spotted in the Archipelago Sea, Gulf of Finland, Bothnian Sea and the Quark. The bloom situation developed rapidly and spatially highly variable, even over short distances. The fragmented nature of the coastal areas and changing wind and water currents makes the algal bloom conditions highly dynamic.

2.2 Main Instruments

To understand the chemical composition of the precursor vapors emitted from various sources around the site, the Chemical ionization Atmospheric Pressure interface-Time Of Flight mass spectrometer (CI-APiTOF) was operated from the 4th floor laboratory of the Physicum building, Kumpula campus,

University of Helsinki (60° 12' N, 24° 58' E; 49m, a.m.sl). The other aerosol and trace gases instruments were operated at the SMEAR III station which is 180 m away from the mass spectrometric measurement site (Station for Measuring Ecosystem-Atmosphere Relation (SMEAR III), 60.20° N, 24.96° E; 25 m a.s.l.).

The Atmospheric Pressure interface-Time Of Flight (APiTOF) mass spectrometer is the state-of-the-art instrument for gas phase chemical composition investigations including aerosol precursor characterizations. Here the instrument is coupled with a nitrate based-chemical ionization (CI) inlet in order to measure neutral gas-phase molecules that are clustered and charged with a reagent ion. In our study we used inlet design as described by Eisele and Tanner (1993) and Kurten et al. (2011) and further used by Jokinen et al., 2012. The Time Of Flight (TOF) mass analyzer can detect molecules with masses up to 2000 Th with a mass resolution of 3600 Th/Th. More details on the working principle of the instrument and calibrations can be found in earlier studies (Junninen et al., 2010, Jokinen et al., 2012; Kürten et al., 2014). The sampled air was drawn in through a 1 m-long, "3/4" diameter stainless steel tube with an average flow rate of 10 Lpm. In this study, the chemical ionization was done via nitrate ions (NO₃-) through X-ray exposure of nitric acid (HNO₃, flow rate: 3 mLpm), saturating the sheath air flow entering the CI (flow rate: 30 Lpm), the inlet flow of 10 Lpm was reached by using a 40 Lpm total flow. The instrument was calibrated prior to the experiment according to (Kürten et al., 2012) resulting in a calibration factor of 1.45 × 10⁹ molecule per normalized unit signal including the diffusion losses in the inlet line.

The resulting data (i.e. obtained signals) were averaged to 60 min before the mass calibration step performed through the MATLAB based program tofTools (Junninen et al., 2010). SA, MSA, IA concentrations are calculated after normalizing them with the reagent ions (NO₃⁻ and (HNO₃)NO₃) using the equation mentioned in Jokinen et al., 2012. The uncertainty range of the measured concentrations reported in this study is estimated to be –50%/+100% and the limit of detection, LOD: 4×10⁴ molecules cm⁻³ (Jokinen et al., 2012). HOMs and IA have been estimated to be charged similarly at the kinetic limit as SA (Ehn et al., 2014; Sipilä et al., 2016), so the calibration factor for them should be similar, but please note, that the concentration of other compounds than SA can be highly uncertain due to different ionizing efficiencies, sensitivities and other unknown uncertainties. If MSA, IA or HOMs do not ionize at the kinetic limit these concentrations could be underestimated and thus, the concentrations reported in here should be taken as low limit values. The normalized signals of specific HOMs found in the study are calculated using high resolution peak fitting data. Please note that the concentration of highly oxygenated molecules (HOM monomers and dimers) were calculated from the unit mass resolution data.

Neutral cluster and Air Ion spectrometer (NAIS, Airel Ltd., Estonia, Manninen et al., 2010; Mirme and Mirme, 2013) was used to measure the number size distribution of both positive and negative ions between 0.8 nm and 42.0 nm (electric mobility diameter). The NAIS also measures the number size distribution of total particles (neutral) between 2.5–42.0 nm. NAIS consists of two multichannel electrical mobility analyzer columns (DMA's) operating in parallel. The columns differ by the polarity of the ions measured, but are otherwise identical (Mirme and Mirme, 2013) in operation. However they may differ in the transfer functions after calibration. The calibration procedure for the DMAs is presented in Mirme and Mirme, 2013. The ion mode measurements are corrected as in Wagner et al., 2016)". The flow rate of the instrument is 60 Lpm which is split into 30 Lpm for each DMA. The instrument was installed in the SMEAR III station. The data was recorded every 2 s.

Larger particles of 3–820 nm were measured using a twin differential mobility particle sizer (DMPS) (Aalto et al., 2001). The instrument was installed in the SMEAR III station. The time resolution of data is 10 minutes.

The size distribution of 1–3 nm particles was measured by a Particle Size Magnifier (PSM, Airmodus Ltd., Finland; Vanhanen et al., 2011) in series with a condensation particle counter (Airmodus Ltd., Finland). The PSM was operated by scanning the flow 0.1–1.3 lpm (continuously changing the saturator flow rate) which allows determining the 1-3 nm particle concentration and calculation of particle size distribution. The data was recorded for each second and the duration of each scan was fixed to 240 s. The raw data inversion was carried out through the kernel method (Chan et al., 2020; Lehtipalo et al., 2014). The raw data of the PSM employed a pretreatment filter that calculates the correlation between the observed particle concentration and the saturator flow rate of a single scan and discards scans with significant non-correlation or negative correlation (Chan et al., 2020). Details about the Back-trajectory calculations, Chla data analysis, meteorological and other calculations of parameters such as growth rates and formation rates are explained in the SI.

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3. Results and discussions

256 3.1 Meteorological parameters and cyanobacterial bloom during the study.

257 3.1.1 Meteorological Parameters

258 The meteorological parameters, especially the wind speed, wind direction and ambient temperature, 259

varied significantly during the study period. The time format in the entire study is UTC+02:00 h. This

260 study period includes the hottest summer days of Finland in year 2019. The average temperature

during 17–28 July (the warmest period) was 21.6° C with a maximum temperature of 31.6° C recorded

on the 28 July (Fig. 2). Temperature starts to decrease after 29 July. The average temperature in August was 16.5° C with a maximum temperature of 21.9° C recorded on 5 August 2019.

The wind direction was highly variable during June-July period. The wind direction in July was mostly from the sectors 270° – 320° (West-Northwest) and 90° – 150° (East-South East). In August, the wind gained more stability and was dominantly blowing from 180° – 270° (South-West) (Fig. 2). The wind speed also showed high variability in June-July. The wind speeds during June and early weeks of July were mostly >6.5 m s⁻¹, followed by a bit calmer mid-July (mostly <=4 m s⁻¹) with preceding high winds in end of July until mid-August (gusts of winds > 5.2 m s⁻¹) (Fig. 2). However, the average wind speeds in both the months was 3 m s⁻¹. The average daylight hours in July were 17-18 hours with the daytime hours between 04:00–22:00 h which starts to decrease in August to 15–16 hours of daylight per day (05:00 h – 21:00 h) as per the Global radiation data obtained from SMEAR III station for the study period. Therefore, the actual nighttime hours in our measurement site can considered from 23:00 h–03:00 h during Finnish summers.

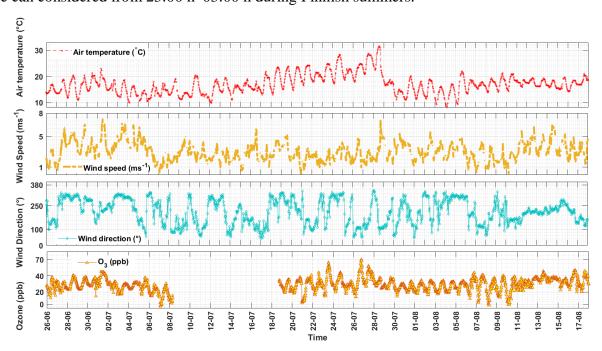


Figure 2: Time series of meteorological parameters and O₃ (data from SMEAR III station, 30-minute averaged) during the study period.

3.1.2 Cyanobacterial bloom conditions during the study

The Baltic Sea (defined from 53° N to 66° N latitude and from 10° E to 30° E longitude inclusive of Gulf of Bothnia, Gulf of Finland and Gulf of Riga) is characterized by usually two algal blooms occurring in early Spring (mostly diatoms) and a summer bloom increasingly dominated by cyanobacteria (blue green algae). The summer bloom period selected for this study was typically

characterized by cyanobacteria. When these microscopic cyanobacteria multiply and aggregate, they are seen as blue-green patches or scum-like layers over the surface of lakes and marine waters. The warm early summer temperatures (during June) resulted in a cyanobacterial bloom (Finnish national monitoring; SYKE press release, 2019). However, the weather conditions in end of July began changing with high winds causing the cyanobacteria to be highly mixed in the water column, which reduced bloom intensity at the sea surface to lower than normal mean cyanobacterial biomass (mean biomass of cyanobacteria, 105 µg L⁻¹, Kownacka et al., 2020) in end of July and August (SYKE press release, 2019). However the average biomass of cyanobacteria in 2019 (196 µg L⁻¹, Kownacka et al., 2020) was slightly higher than the average. Subsequently, temperatures were lower in August as compared to June and July and windier as compared to other summer months. These windy conditions kept the lake cyanobacteria well mixed in the water. The northern Baltic Sea, including the Gulf of Finland, the Southern parts of the Åland islands and even the Bothnian Sea occasionally observed massive blooms of cyanobacteria during June-August 2019. However, the bloom intensity of cyanobacteria at the coastal areas were intermittent and changed rapidly due to the spatial complexity of the coastline and variable winds and currents.

These cyanobacterial blooms are generally dominated by three taxa, Nodularia spumigena, Aphanizomenon sp. and Dolichospermum sp. (Knutson et al., 2016; Kownacka et al., 2020). In the Baltic Sea, these cyanobacteria actually contribute the most to the total pelagic nitrogen fixation (Klawonn et al., 2016). Other potential primary producers emitting vapors are the littoral macroalgae growing along the shallow coastline. For example, the perennial macroalgae, Fucus vesiculosus covers large areas of the coastal areas of Baltic Sea, where they support very high biomass and high productivity (Attard et al., 2019). Low sea levels (0.2–0.8 m, wave height at Suomenlinna aaltopoiju station, https://en.ilmatieteenlaitos.fi/wave-height) were recorded in mid-July (11 July 2019–27 July 2019) during the period when high temperatures (20° C and above) prevailed (Fig.2) in our study region. During these conditions, contributors to emissions might be a mix of both coastal macroalgae and open sea microalgae, which are mostly the cyanobacteria. There is a possibility that reasonably, large extents of coastal macroalgae, including F. vesiculosus, were exposed to direct sunlight (in shallow waters or low tide conditions) during the decay of the blooms during mid-August (when the bloom intensity was low, SYKE press release, 2019), hence making this time window favorable for observing potentially high emissions in gas phase from macroalgae, in addition to the emissions from cyanobacterial blooms. However, in the semi-urban/coastal setting of this measurement site, there could be various other parameters, which also could play a role in determining the concentrations of the biogenic emissions; for example the wind speed and wind direction. The atmosphere in this semi-urban coastal location is itself a cocktail of various vapors,

oxidants and particles, which would affect the quantification, source apportionment and characterization of the biogenic emissions.

The measured daytime precursor vapor concentrations showed a regular diurnal cycle consistent with

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3.2 Precursor vapor concentrations and their sources

323 the photochemical production of SA and IA in 90% of the days in this study. SA, key precursor of 324 atmospheric NPF, is formed mainly by reaction of sulphur dioxide with OH-radicals, which is predominantly controlled by the photochemical cycles (Sipilä et al., 2010; Jokinen et al., 2017). The 325 daily mean concentration of SA in July and August were almost similar, 3×10^6 molec. cm⁻³. The 326 mean concentration is slightly lower as compared to the concentrations of SA measured in Helsinki 327 street canyon, 1×10⁷ molec. cm⁻³ (Olin et al., 2020) but similar to the SA concentration measured at 328 329 the SMEAR III station in 2018 (Okuljar et al., 2021). In the study of Olin et al., 2020, SA 330 concentrations were greatly affected by vehicular traffic as the site is situated at a busy street canyon. 331 The SMEAR III is considered as a background site much less affected by vehicular traffic (Okuljar 332 et al., 2021). In comparison to other locations, the daytime SA concentration in pristine Antarctic region has been reported from 10⁵ up to 10⁷ molec. cm⁻³ (Mauldin et al., 2001, Jokinen et al., 2018), 333 10⁶ molec. cm⁻³ in remote continental, remote marine and forest regions and 10⁷ molec. cm⁻³ in urban 334 and rural agricultural lands using the same technique as in here (Berresheim et al., 2000; Kuang et 335 336 al., 2008; Petäjä et al., 2009; Kurtén et al., 2011; Zheng et al., 2011; Chen et al., 2012; Jokinen et al., 337 2012; 2017, Kürten et al., 2014; Bianchi et al., 2016; Baalbaki et al., 2021; Dada et al., 2020). It has 338 been well documented that SA contributes to aerosol formation and growth processes (Boy et al., 339 2008; Eisele et al., 2006; Fiedler et al., 2005; Iida et al., 2008; Sarnela et al., 2015; Jokinen et al., 340 2018; Kürten et al., 2015, 2016; Mauldin et al., 2001; Paasonen et al., 2010; Wang et al., 2011; Weber 341 et al., 1998, 1999; Yao et al., 2018; Dada et al., 2020). Most of these studies are conclusive that SA 342 concentration in the atmosphere depends on the anthropogenic and biogenic activities around the site. 343 In the coastal marine boundary layer, the MSA concentration is typically 10–100% of 344 that of SA (Berresheim et al., 2002; Eisele and Tanner, 1993). Until recently, no studies have been 345 found to report MSA and IA concentrations in coastal/urban setting of Finland. The daily mean concentration of MSA in July and August was almost similar, 4×10⁵ molec. cm⁻³. The mean 346 concentration of IA in July and August was 1×10^6 molec. cm⁻³ and 3×10^6 molec. cm⁻³, respectively, 347 348 showing two times increase in IA concentrations in August (Fig. S1). A similar increase in IA 349 concentrations from summer to autumn were observed in the Arctic Ocean, where the increase in IA 350 was attributed to the freezing onset of the pack ice and increase in ozone concentrations (Baccarini et 351 al., 2020). However, here the increase is mainly due to the change in the air mass arriving at the

experimental site, enriched with biogenic emissions from the blooms. For the same period, the CI-APiTOF data shows exceptionally high concentrations of highly oxygenated organic molecules (HOMs), with monomer concentrations (300–450 amu) of 10⁸ molec.cm⁻³ and HOM dimer concentrations (450–600 amu) of 10⁸ molec.cm⁻³ as well (Fig.S2).

The IA concentration rises one order of magnitude, from 10⁶ to 10⁷ during the 11–17 August, when the wind direction changes abruptly (from 280°–360° to 180°–230°, marine air mass, Fig. 3). We found that during the marine air (180°–230°, South Easterly, over Gulf of Finland and South westerly, over Northern Baltic sea) influence over the study region the average noontime maximum of SA, IA is on the order of 10⁷ molec.cm⁻³ and MSA is around 10⁶ molec.cm⁻³ (Fig. 3). This is one order of magnitude higher concentration than when the wind was from over land (Fig. 3).

The highest concentration, 3×10^7 molec. cm⁻³ of IA was observed when the wind is coming from the Baltic sea sector, whereas the highest SA concentrations ($^{\circ}3 \times 10^7$ molec. cm⁻³) was observed when air mass travelled over the countries of Estonia and Russia crossing Gulf of Finland before entering the measurement site (land+sea region). The connection between the aerosol precursors and the wind direction can be observed in the cases where the wind direction changes rapidly. The highest IA concentration was recorded when the wind direction changes after the 4 August, $180^{\circ}-230^{\circ}$ (the Baltic Sea region). The change in wind direction was clearly reflected in a reversal of the concentration trends of SA and IA (Fig. 3). It was observed that the winds coming from $80^{\circ}-180^{\circ}$ or $250^{\circ}-280^{\circ}$ (land-sea region, Fig. 3) were SA rich air masses. This comprises of the landmasses of south and northeastern Finland, Northern Russia, part of Gulf of Finland and Estonia and North-North western part of Finland including a part of northernmost Gulf of Bothnia. The sector $0^{\circ}-90^{\circ}$ or $280^{\circ}-360^{\circ}$ (land, Fig. 3) consists mostly of urban cities.

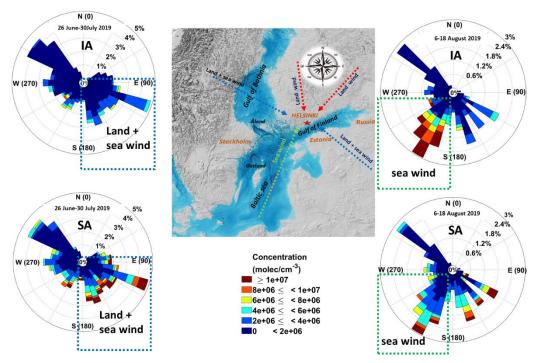


Figure 3: Windroses showing the variability in the concentration of gases with wind direction during the study period. Percentages on the concentric circles denote the frequency of winds from different directions. The spokes are color coded as per the concentration of the gas from the particular direction. The numbers in the parenthesis within the windroses refer to the wind direction in degrees.

During the entire study period, when the air plume passed over the northern Baltic Sea region and the wind speed was high enough (> 4m s⁻¹) high concentrations of IA was observed. While IA can be exclusively sourced from the marine and biogenic emissions (Mahajan et al., 2011; O'Dowd et al., 2002; Sipilä et al., 2016; Carpenter et al., 2021), SA could be biogenic or /and anthropogenic. Further, the temperatures prevailing during this period may have facilitated the DMS oxidation at a higher rate, which forms the source of biogenic SA and MSA. However, this is not a very simple equation, since this fractional yield of biogenic SA from DMS oxidation additionally also depends on the atmospheric $NO_x(NO + NO_2)$ and $HO_x(OH + HO_2)$ levels and on the scavenging of SO_2 by sea salt or cloud droplets (Hoffmann et al., 2016). The anthropogenic sources of SA for this site could also include vehicular or ship traffic especially considering that there is a city road just 250 m and a harbor 6 km away from the measurement site. We explored the correlations of SA to a biogenic proxy, MSA and correlation with NO_x (anthropogenic proxy) to have a clear source apportionment of SA (Fig.4). SO_2 could not be treated entirely as anthropogenic proxy as it can be sourced from DMS oxidation as well.

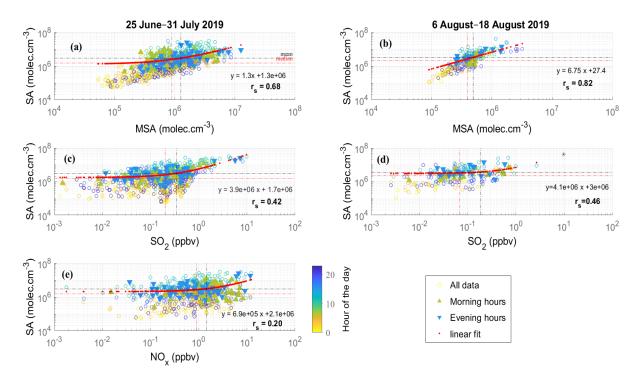


Figure 4: Correlation of SA with MSA (a,b), SO_2 (c,d) and NO_x (e) for June–July. The black dashed lines for both axis represent the mean of the gas concentration, red dashed line represent the median value the gas concentrations and red solid line represents the linear fit. Spearmann's coefficient (r_s) was used to test the correlation, at significance level, 0.001. The circles represent data points at different hours of the day. The upward pointing green triangles represent the morning rush hours (6:00–8:00 h) and the downward pointing blue triangles represent the evening rush hours (15:00–17:00 h). The yellow hollow circles represent all data. NO_x data unavailable of August.

The good correlations ($r_s > 0.6$, Fig. 4a and 4b) between SA and MSA during the study period (June–August) could suggest that they were sourced from a common biogenic source, the DMS emission from the cyanobacterial bloom. Good correlations of SA and MSA was also found in August ($r_s = 0.8$, Fig. 4b) when the air mass was mostly marine (and/or from the Finnish coastline, Fig. 3). Another observation was that SO_2 also shows some correlations with SA in both June-July and August study periods ($r_s = 0.4$, Fig. 4c and 4d), but not as significant as SA and MSA correlations. SO_2 can have different sources unlike MSA which is mostly biogenic. However some emissions could be sourced from agriculture and other terrestrial sources, Bates et al., 1992, hence these observations could possibly indicate SA was more biogenic than from other sources. But we cannot be very accurate in this estimation only by analyzing the correlation coefficients since both MSA and SA can have a similar daily cycles due to the oxidation pathways.

Both SO₂ and MSA are the oxidation products of DMS (produced by phytoplanktons, including some cyanobacteria), oxidized through OH and NO₃ radical (Chen et al., 2000). Some of the previous chamber studies have confirmed that SO₂ is the major intermediate products formed from DMS oxidation (Sørensen et al., 1996; Berresheim et al., 1995). The SO₂ could be oxidized to SA (OH/O₂) oxidation) during the transport. Since our experimental site was surrounded by water bodies and the summer season had enriched most of these freshwater and marine waters with abundant cyanobacterial blooms, this biogenic SA contribution to the study site has to be accounted when analyzing the sources of SA. However, SO₂ can also be sourced from various anthropogenic activities and can be oxidized to SA. In Finland the major sources of anthropogenic SO₂ is the public power industries contributing to almost 90% to the total SO₂ emissions in Finland in the year 2019, while transport contributing to less than 1% according to the emission inventory prepared by Finnish (Finnish Pollution Environment Institute, **SYKE** Air Inventory; ymparisto.fi/en-US/Maps_and_statistics/Air_pollutant_emissions). Further the maximum data points of high concentrations of SO₂ (~10⁷ molec. cm⁻³) were not observed during the traffic hours in June-July-August (Fig. 4c and 4d) another possible indication that biogenic sources could be contributing to the SO₂ concentrations and thus SA concentrations near the study site.

The emission inventory of Finland for the year 2019 indicated that sources of NO_x as NO₂ were mainly the power industries (41.5%) and the transport sources (41%) (ymparisto.fi/en-US/Maps_and_statistics/Air_pollutant_emissions). These sources are indeed the most significant sources of NO_x globally (Meixner and Yang, 2006). NO_x, definitive proxy of anthropogenic influence shows a poor correlation with SA (r_s=0.28, Fig. 4e) during June-July which could suggest insignificant effect of traffic on the SA concentrations. Unfortunately, the NO_x data from August was unavailable due to instrument malfunction so we cannot provide any correlations for this month.

The data presented in Figure 3, where we observe high SA concentrations even when the air mass was marine and the good correlations of SA-MSA (inclusive of insignificant correlations of SA-NO_x) (Fig. 4) indicate towards a greater possibility of the influence of biogenic emissions on the concentrations of SA as compared to the anthropogenic emissions.

3.3 Types of nucleation events during the study

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During, 25 June 2019–19 August 2019, we observe a number of NPF events characterized by a short appearance of ultrafine particles in the number size distribution lasting for less than one hour. These so-called bursts/spikes appearing at small sizes (sub-3 nm) are indicative of local clustering processes in contrast to regional events, where it is possible to follow the growing particle mode for several

hours (Dada et al., 2018; Dal Maso et al., 2005). Local clustering here means that the molecules could be transported from elsewhere but the actual clustering could have taken place near the experimental site, indicated by a small bump of clusters (with absolutely little or no growth) as seen in the NAIS spectra. We do observe transported events (events with a growing particle mode, but no small particles forming at the site) and non-events days but they are not included in the analysis. This section discusses the occurrence of local and regional new particle formation events with the focus on: 1) trace gases variability during the event days, 2) the evolution of different sized particles during these events, 3) the impact of meteorological parameters and 4) the effect of cyanobacterial bloom on the events.

Table 1: Timing and maximum concentration of SA, MSA and IA during local and burst/spike nucleation events during the study period

Dates	Type of	time of NPF	SA (max)	MSA (max)	IA(max)
	Event	(UTC+02:00	molec. cm ⁻³	molec. cm ⁻³	molec. cm ⁻³
		h)			
30.06.2019	Regional/	8:45-13:23	7.9×10^6	5.6×10 ⁵	2.3×10^{6}
	local	14:00-16:30			
30.07.2019	Regional/	7:45 -11:16	1.2×10^7	1.2×10^6	5.3 ×10 ⁶
	local				
11.08.2019	Ion Burst	13:40-14:32	1.0×10^7	1×10 ⁶	3.2×10^{7}
	(Spike)				
14.08.2019	Ion Burst	8:00-8:20	4.2×10^{6}	5.3×10^5	8.5×10^{6}
	(Spikes)				
15.08.2019	Multiple Ion	6:00, 8:58,	6.4×10^{6}	5.8×10^5	2.5×10^{6}
	Bursts	14:00-16:00	6.3×10^{6}	4.6×10^{5}	3.1×10^{6}
	(Spikes)		7.0×10^{6}	6.8×10^{5}	1.5×10^6

3.3.1 Nucleation: Regional and Local events

A regional NPF event was observed on 30 June 2019, which starts at 08:45 h and ends at 13:23 h (Fig 5a). The negative ion clusters start to increase in concentration first at 08:45 h (Fig. 5b) concurrent with the increase in concentration of the smallest particles (<3nm) from 10^2 to 10^3 cm⁻³ (Fig. 5c). Preceding the NPF event the SA concentrations were steadily increasing and subsequently at 09:00 h, SA concentration doubles from 2×10^6 to 4×10^6 molec. cm⁻³ (Fig. 5d), while the particle formation

rate at 1.5 nm ($J_{1.5}$) increasing from 0.3 cm⁻³ s⁻¹ to 0.6 cm⁻³ s⁻¹. $J_{1.5}$ was much higher than either of $J_{+1.5}$ and $J_{-1.5}$, thus indicating a neutral formation pathway rather than ion mediated. Further we also observe local clustering event at 15:00 h with simultaneous increase of concentration of SA and HOMs along with increase in the smallest particle concentration. This possibly indicates the role of SA and HOMs in the nucleation initiation. The high normalized signals of DMA-SA cluster seen during the entire event (increasing from the start of NPF event) possibly indicates that SA clusters initiate the event (Fig. S4a). DMA inclusive of other main methylamines like mono and tri methylamines (Bergman et al., 2015) in the global inventory (Schade and Crutzen, 1995) is contributed through the animal husbandry and other agricultural practices, biomass burning and some contributions from marine and terrestrial sources. Although among these methylamine emissions, generally the trimethylamine dominates (Schade and Crutzen, 1995). Although no estimates of DMA measurements are available from Helsinki region, the DMA in a boreal forest site in Finland has been estimated to be below ~ 150 ppqV (Sipilä et al., 2015), measured through a NO3- CIMS. Their work also stated that DMA was unlikely the playing an important role in the nucleation process observed at the site.

The increase of HOMs is also clearly observed during the event Fig. S4b. Therefore we suggest that nucleation and growth of particles was possibly due to SA-organics which ensures that particles reach the CCN and thus climate relevant diameters. The work of Okuljar et al. (2021) also report an increase in sub-3 nm particles with a simultaneous increase in SA concentration at the SMEAR III site, supporting our observations. However, the role of HOMs in nucleation initiation has not been explored at this site.

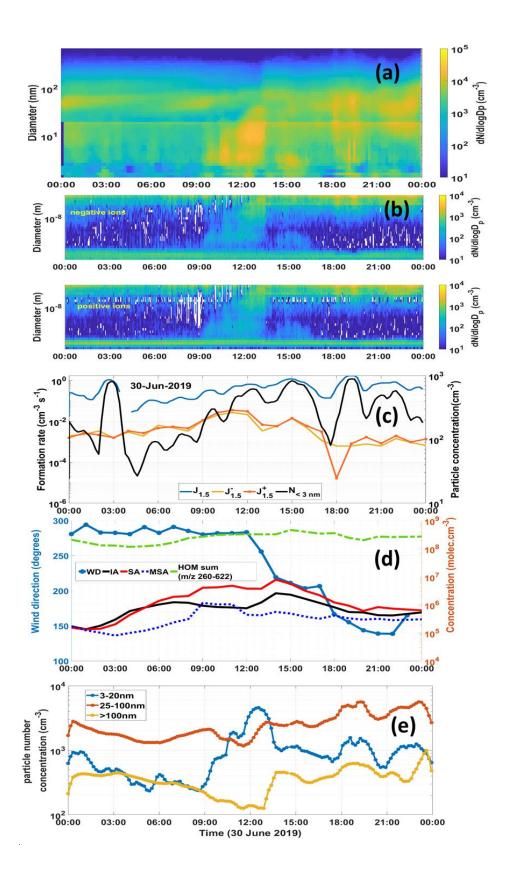


Figure 5: NPF Event (Regional and local events), 30 June 2019. (a) Number size distribution of particles (data from PSM, NAIS and DMPS; size range: sub-3–100nm). (b) Charged particles number size distribution (negative: upper, positive: lower) obtained from the NAIS. (c) Diurnal variation of

formation rates ($J_{1.5}$) of 1.5 nm particles and ions ($J_{1.5}$ and $J_{1.5}$) on the left axis and particle number concentrations (1.5–3 nm) on the right axis. (d) Diurnal variation of HOMs SA, IA and MSA with wind direction (WD). (e) The diurnal variation of particle concentration in nucleation:3–20 nm; aitken: 25–100 nm and accumulation: >100nm) mode particles during the event (Data from DMPS).

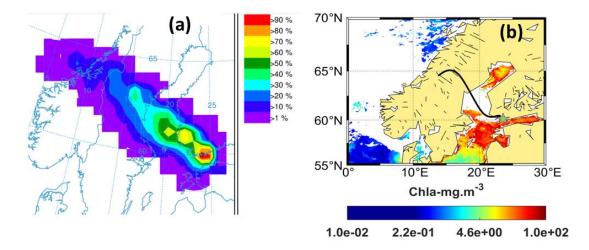


Figure 6: (a) Trajectory frequency plot (100 a.g.l, arrival time of trajectories at the meaurement site: 20:00 h) for 24 h back trajectory using GDAS meterological input data (frequency grid resolution: $1.0^{\circ} \times 1.0^{\circ}$) (b) Chl-a concentrations (GlobColour level-3); Black line shows the trajectory direction and the star point denotes the measurement site.

A clear increase in nucleation mode particles is seen during the event, starting at 08:45 h (234 cm⁻³) and reaching its maximum at 12:30 h (4589 cm⁻³). This increase in concentration of the nucleation mode particles was followed by the increase in concentration of Aitken mode and accumulation mode particles and continues for a couple of hours, indicating growth of particles (Fig. 5e), possibly reaching to CCN relevant sizes. However, we also observe a drop in Aitken particles before NPF which also continues during NPF. We speculate it could be due to the change in wind direction (Väkevä et al., 2000) before NPF. The wind direction relatively remains constant throughout the NPF so the low concentration of Aitken mode continues. Wind direction changes abruptly at 12:00h and the Aitken mode particle concentrations increases soon after this change of wind direction (Fig. 5d). This shows the particles must be the process of growth mostly elsewhere, which is not evident in the changed air mass, however we still observe almost the same (or even slightly higher) precursor vapor concentrations, since the wind still passed over the bloom areas before entering our study site. After the change in local wind direction, the observed SA and IA slightly increase, and we still observe clustering (formation of small ions and particles), but no continuous growth typical for regional

events. Figure 6a shows that >40% of the trajectories passes above the Swedish island of Gotland towards southern part of Bothnian Sea. The satellite data shows that the bloom was present in the Bothnian Sea, but not quite dense as compared to the southern Baltic Sea (south of Gotland island) and the northern part of the Gulf of Finland (Fig. 6b). The majority of the trajectories did not pass over the dense cyanobacterial bloom patch during this day (Fig. 6b). The calculated normalized residence time was higher over the neighboring cities of Helsinki (Southwestern side) and parts of Bothnian Sea during the event time (see Fig. S3). Thus the land based anthropogenic activities and biogenic sources both can be contributing to SA concentrations for this event; here we cannot exactly quantify the source types for SA. However, the source of SA from the local sources such as vehicular traffic around our measurement site is small (as discussed above) but cannot be completely ignored (Olin et al., 2020).

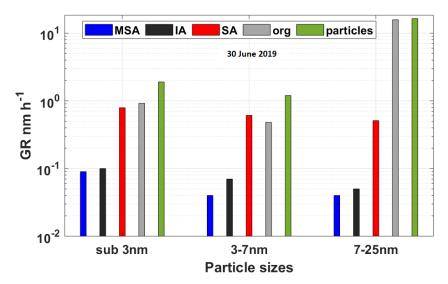


Figure 7: Particle growth rates calculated from the kinetic condensation of gases (data from CI-APi-ToF) and the measured particle GRs (data from NAIS) in different size classes on 30 June 2019.

The particle GR (7–25 nm) for this event was 16.5 nm h⁻¹, which is typical of a coastal site. Even when several condensing vapors participate in the growth process, growth rates typically do not exceed 20 nm h⁻¹ (Kulmala et al., 2004). The GR for organics was calculated after subtracting the combined contribution of the GR of SA, IA and MSA from the measured particle GR (Fig. 7). The GR for organics should be treated as an estimation since no separate GR calculations and assumptions were used. The calculated growth rates (GR) shows that SA can explain maximum 41% of the growth of sub-3 nm particles, while IA and MSA can explain only <1% of the GR in this size range. The GR by SA in the bigger size fraction (7–25 nm) was only 0.51 nm h⁻¹ explaining only 3% of the measured growth rate of particles. This means that vapors other than SA, IA and MSA were

responsible for 96% of the measured particle growth. These other vapors could include different organics since organics are known to contribute to growth of particles (Kulmala et al., 1998, 2004; Riipinen et al., 2012; Zheng et al., 2020) and explain particle growth in the boreal forest (Ehn et al., 2014).

Another example of regional event (neutral nucleation) probably driven by SA and organics was observed on 30 July 2019 (Fig. S5) which lasts for around four hours. The trajectory frequency plots showed that most of the trajectories were from the northern land areas (including urban cities and boreal forests) of Finland with highest residence times over these land regions (Fig. S6 and S7). Since, the precursor gases from the biogenic origin, IA and MSA, do not show a significant concentration increase as compared to SA, at the start of the event, their contribution towards the initiation of the NPF event may not be as significant as SA. The greater residence times over the land areas clearly support the high SA and organic concentrations seen during the event indicating a SA driven event – with a possible contribution of HOMs (Fig. S7). In this case, SA explains 60% of growth of sub-3nm particles compared to 41% when the dominating trajectories passed over the Gulf of Finland (Fig. 5, 30 June 2019). Still, as for the previous case, a major fraction of the growth in the 3–7 nm range remains unexplained by the available acids (SA, IA, MSA) and is expected to be related to the contribution of organics. The GRs explained by SA in both sub-3 nm (1.93 nm h⁻¹) and 3–7 nm (1.46 nm h⁻¹) size ranges are 58-59% higher than on 30 June 2019 (0.79 nm h⁻¹ and 0.61 nm h⁻¹ for sub-3 nm and 3-7 nm, respectively) which could be explained by the increase in SA by 52% on 30 July 2019. Thus, the events on 30 June and 30 July possibly occur via the nucleation of SA (possibly stabilized by bases eg. ammonia or amines) and the HOMs contribute to growth of particles and possibly in nucleation as well.

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3.3.2 Nucleation: Burst events

Case 1: Biogenic IA nucleation-burst/spike events, 11 August.2019

567 Intense burst events are frequently observed at coastal sites accompanied with high concentrations of 568 IA (O'Dowd et al., 2002; Rong et al., 2020; Sipilä et al., 2016). Two of such bursts or spike events 569 were observed on 11 August 2019 at 04:00 h and 13:00 h (Fig. 8a). Only the second spike event was 570 observed in the NAIS size distribution with a higher intensity in the negative mode at 13:00 h (Fig. 571 8b). During both these spike events we observe the formation of clusters (1.5 nm) and the formation rate ($J_{1.5}$) increases from 0.2 to 3.7 cm⁻³s⁻¹ during the event with a simultaneous significant increase 572 in the sub-3 nm particle concentrations from ~100 to >2000 cm⁻³ (Fig. 8c). J_{L5}^+ and J_{L5}^- remain lower 573 574 than the total formation rate indicating this event to be a case of neutral nucleation. At the same time, IA shows increase in concentration from 9×10⁵ molec. cm⁻³ at 03:00 h to 1×10⁶ molec. cm⁻³ at 04:00 575

h. During this event the air masses changes from 160° to 140° i.e the direction of the airmass is changed to the Gulf of Finland. In the second burst (at 13:00 h), the IA concentration increases from 2×10^6 to 7×10^6 molec. cm⁻³ from 13:00 h to 14:00 h (Fig. 8d) with a slight change in wind direction from 151° to 166° Most of these air masses are from the Gulf of Finland. SA concentration also increased but remained lower than IA during both the burst/spike events indicating a possibility that iodine oxoacid formation initiates cluster formation (He et al., 2021). We observe a growth of particles until 15:00 h in the particle modes (NAIS data, Fig. 7b). However the particles are seen reaching sizes up to size 100 nm (DMPS data, Fig. 8a). The organics almost remain constant within the range of $2-3 \times 10^8$ molec. cm⁻³. A further increase in IA concentration, 3×10^7 molec. cm⁻³ occurs at 15:00 h, and the concentration remains in the range of 10⁷ molec. cm⁻³ for another two hours (Fig. 8d). This was the highest observed IA concentration in the entire measurement period. A recent study by He et al., 2021, indicate that IA concentrations above 1×10^7 molec. cm⁻³ leads to rapid new particle formation at +10° C. At such concentrations the efficacy of iodine oxoacids to form new particles exceeds that of the H₂SO₄-NH₃ system at the same acid concentrations. Thus, the concentration of IA found in this event (two times higher than SA during the start of the event), the high formation rates (>1 cm⁻³ s⁻¹) and an unchanged concentration of SA during the event, as compared to the event on 30 June 2019, strongly suggests that it could be an IA driven-NPF event.. In addition, a clear increase in the normalized signal of deprotonated IO₃ with no significant increase in DMA-SA cluster during the event at 13:00 h (Fig. S8a). However, HNO₃-IO₃- cluster was the most abundant followed by the H₂O-IO₃⁻ cluster indicating this event to be IA-driven nucleation. Further, between 14:00-15:00 h, when we observe the highest IA concentrations a subsequent growth of particles is noted. We also observe an increasing number concentration of nucleation mode particles from 13:40 h (~650 cm⁻³) to 14:40 h (~1800 cm⁻³). After this one hour of intense clustering, the Aitken mode particles also begin to increase in concentration from ~1300 cm⁻³ to ~4800 cm⁻³ during 15:00 h–18:00 h (Fig. 8e). The total particle concentration increased from ~2400 cm⁻³ to ~6400 cm⁻³ within an hour during this burst event. We suggest that this burst event was possibly capable of producing particles big enough to act as CCN. Since it was an intense burst event with no proper horizontal growth (as seen in "banana" type events), we were not able to calculate the growth rate for this particular event. Therefore we are unable to quantify the contribution of IA towards the growth of particles reaching CCN sizes.

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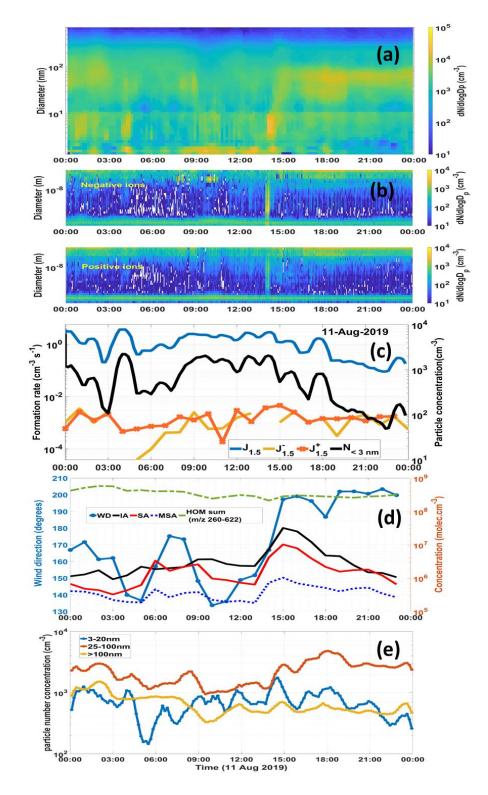


Figure 8: Burst/Event, 11 August 2019. (a) Number size distribution of particles (data from PSM, NAIS and DMPS; size range: 1–100 nm). (b) Charged particles number size distribution (negative: upper, positive: lower) obtained from the NAIS. (c) Diurnal variation of formation rates ($J_{1.5}$) of 1.5 nm particles and ions ($J_{1.5}$ and $J_{1.5}$) and total number concentrations of particles (<3 nm, PSM). (d) Diurnal variation of HOMs, SA, IA and MSA with wind direction (WD). (e) The diurnal variation of

particle concentration in nucleation (3–20 nm), Aitken (25–100 nm) and accumulation mode (>100 nm) particles (DMPS data).

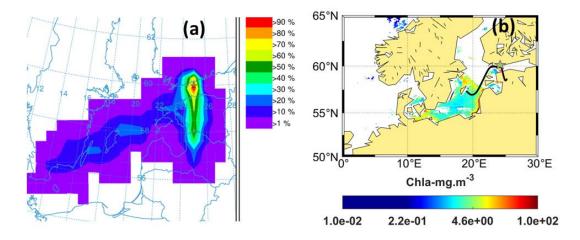


Figure: 9 (a) Trajectory frequency plot (100 a.g.l, arrival time of trajectories at measurement site: 22:00 h) for 24 hour back trajectory using GDAS meteorological input data (frequency grid resolution: $1.0^{\circ} \times 1.0^{\circ}$) (b) Chl-a concentrations (GlobColour level-3); Black line shows the trajectory direction and the star point denotes the measurement site.

The global radiation and brightness parameter suggest that 11 August 2019 was an overall a cloudy day until 12:30 h (Fig. S9). The weather starts to turn into clear-sky after 13:00 h when the brightness parameter increases from <0.3 to ~0.7 (Fig. S9). Impact of brightness parameter on NPF is also observed in a previous study (Dada et al. 2017). The clearing of the sky could explain the intense spike at 13:00 h in the particle number size distribution as well as in the acid concentrations. For this particular case, we investigated further the source of such high IA concentrations and we found that during this day, the cyanobacterial bloom was observed in three intense patches in the central Baltic sea, southern Gulf of Finland (ship transect route between Helsinki and Tallinn) and Gulf of Riga (Fig. 9b). The trajectory frequency analysis clearly shows that the maximum frequency of trajectories was observed over southern Gulf of Finland (inclusive of the coastal waters of Suomenlinna island) however we do see the air masses coming in from the central Baltic sea as well which was characterized by intense bloom during this day (Fig. 9a). The sea level was also low as it was observed to be 0.8–0.9 m in the coastal waters in around the measurement site (Suomenlinna and Gulf of Finland coastal measurements sites), supporting the exposure of the macro to sunlight which can be a good source of iodine precursors.

The residence time of the airmasses coming from the Gulf of Finland and Northern Baltic Sea were longer than the residence time of the airmasses coming from the neighboring land areas (Fig. S10) clearly explaining the source of high IA observed during the event, which is through the blooms. Further, the airmass was completely marine at 15:00 h when the highest IA is recorded supportive of the marine biogenic source of IA and its transport to the measurement site. The distance from the Gulf of Finland to the measurement site is approximately 5-10 km. With the wind speed of 5 m s⁻¹ recorded during the event, it takes less than one hour for the emission to transfer to our measurement site. By the time the air mass reached our measurement site from the emission source, a fraction of the emitted I₂ could have oxidized to IA. However, at this point we cannot differentiate between the sources of IA from neighboring coastal waters and the central Baltic Sea but can speculate that most of the IA observed could be sourced from the nearest coastal locations of Gulf of Finland.

Another burst/spike event driven by IA occurred on 14 August 2019 (Fig. S11) when the IA concentration was found to be 8×10^6 molec.cm⁻³ which was 2 times higher than SA concentration (4×10^6 molec.cm⁻³). The event did not last more than 30 minutes. The precursor vapor concentration was not large enough for the event to continue or the particles to grow further. The meteorological conditions were very much similar to this event (11 August 2019). For this event also, the airmass was marine with maximum residence times over the Gulf of Finland and Baltic Sea regions. Vicinity of the emissions to the measurement site enabled the detection of these fast-forming clusters.

Case 2: Biogenic SA nucleation –multiple bursts events

Another kind of event was observed on 15 August 2019 (Fig. 10a) where multiple particle bursts are observed and the particles grow to sizes > 50 nm.

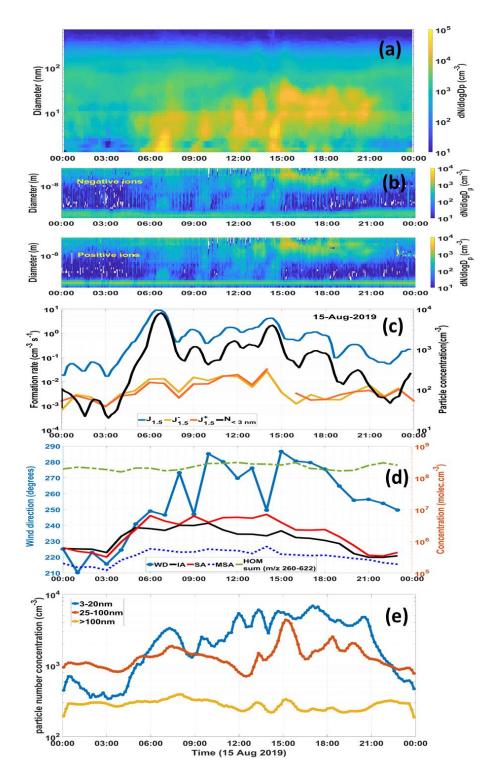
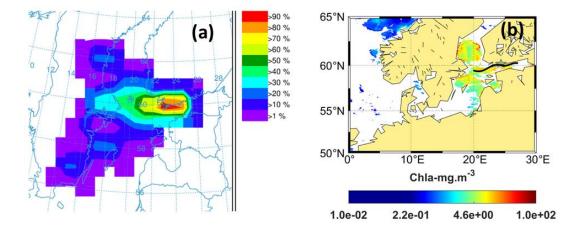


Figure 10: Multiple Burst/Spikes, 15 August 2019. (a) Number size distribution of particles (data from PSM, NAIS and DMPS; size range: 1–100nm). (b) Charged particles number size distribution (negative: upper, positive: lower) obtained from the NAIS. (c) Diurnal variation of formation rates ($J_{1.5}$) of 1.5 nm particles and ions ($J_{1.5}$ and $J_{1.5}$) and total number concentrations of particles (<3 nm, PSM). (d) Diurnal variation of HOMs SA, IA and MSA with wind direction (WD). (e) The diurnal

variation of particle concentration in nucleation (3–20 nm), Aitken (25–100 nm) and accumulation mode (>100 nm) particles (DMPS data).



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Figure 11: (a) Trajectory frequency plot (100 a.g.l, arrival time of trajectories at the measurement site: 22:00 h) for 24 h back trajectory using GDAS meterological input data (frequency grid resolution: $1.0^{\circ} \times 1.0^{\circ}$) (b) Chl-a concentrations (GlobColour level-3); Black line shows the trajectory direction and the star point denotes the measurement site.

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The formation rates for the smallest clusters for both the polarities were the same $(J_{1.5}^+)$ and $J_{1.5}^-$ (Fig. 10b and c). This was also a case of neutral nucleation as inferred from the relatively high (as compared to ions) J_{1.5} (neutrals). On 15 August there was a sudden change of wind direction from the 180°– 215° (prominent wind direction during 11–14 August) to 280° and a series of bursts is triggered with the intense formation of clusters (<3 nm) at each burst (Fig. 10d). The two most intense burst events) were associated with an increase in SA from 2 to 6×10^6 molec. cm⁻³ at 06:00 h, and 5 to 7×10^6 molec. cm⁻³ at 14:00 h (Fig. 10d). A third burst at 09:00 h showed an increase in SA from 3 to 6×10⁶ molec. cm⁻³ at 09:00 h interestingly with IA_{max}: 3×10⁶ molec. cm⁻³. In all the three bursts a simultaneous increase in IA and MSA from 03:00 h to 12:00 h is observed, but the SA concentration was two to three times higher than IA and four to five times higher than MSA concentrations. The most intensive burst was at 14:00 h (as compared burst to 6:00 h) when the SA was 3 times higher than IA. This burst was associated with a significant increase in Aitken mode particle concentration (from 1490 at 14:00 h to 4300 cm⁻³ at 15:00 h). The increase in accumulation particle concentration was seen just after one hour from the start of the bursts for both events (06:00 h and 14:00 h). However the increase in accumulation mode particle concentration for these two events was not very significant (~100cm⁻³) although particles reaching a size more than 80 nm (CCN relevant sizes) was observed.

693 We saw DMA-SA clusters during the event (Fig. S12) which supports the observation that this a SA-694 driven NPF event.

During both these events (in fact, all the smaller burst events observed during this day), the trajectories were originating from Sweden (24 h prior to arrival). However, before entering the measurement site the trajectories passed over the Southern part of Gulf of Bothnia and the trajectory frequency was >70% when the wind passed over the cyanobacterial bloom region (Fig. 11a and b). To confirm our findings we checked a day where there was less intense bloom in the Gulf of Finland and Northern Baltic Sea and the dominant airmass did not pass over the bloom patch in Gulf of Finland (Fig. S13) before entering our experimental site. We did not observe an NPF event on this day, thereby suggesting that the airmasses passing over the bloom patches before arriving at our study site were capable of bringing in biogenic precursor vapors capable of initiating NPF events.

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3.4 Possible contributions of biogenic emissions to Precursor gaseous vapors

706 Assuming insignificant anthropogenic SA contribution as discussed in section 3.2, we investigated the other possible sources of SA by evaluating the type of algae present in the water bodies from 708 travelled during the events. The air masses marine algae 709 dimethylsulfoniopropionate (DMSP), which is capable of forming DMS, which subsequently 710 oxidizes into SA and MSA. While very few cyanobacterial species are capable of producing DMSP (Karsten et al., 1996; Jonkers et al., 1998), and its concentration can vary considerably from one 712 species to another (Keller et al., 1989). Moreover, blooms could be well-mixed with other algal 713 species (ESA report, 2000) which are capable of producing DMSP. A recent experiment identified 714 Aphanizomenon as the only cyanobacteria producing DMS (Steinke et al., 2018). The Gulfs of 715 Bothnia and Riga are dominated by the genus Aphanizomenon (Kownacka et al., 2020). In addition, 716 the Bothnian Sea and Gulf of Finland were found to be rich in cyanobacterial genera of 717 Aphanizomenon along with Nodularia and Dolichospermum (Kownacka et al., 2020). As per the 718 previous studies which were carried out as part of the Baltic-wide monitoring (Kowancka et al., 2020 719 and the references mentioned therein), bloom composition is fairly consistent for different regions 720 and seasons from year to year, which makes it possible for us to make close estimations of the species present during our study in a particular region (from where the airmass travels and the residence time 722 over a particular region).

A recent study also indicated that the abundance of DMS producing cyanobacteria, Aphanizomenon has increased in the Bothnian Sea due to decreasing salinity (Olofsson et al., 2020). Moreover, marine waters themselves are a large source of DMS (Kettle and Andreae, 2000) explaining the contribution of biogenic SA in the above-mentioned burst events (15 August 2019). Hence to conclude, the marine regions surrounding the experimental site could be potential sources of biogenic SA. Moreover, high iodine emissions could be expected over the Baltic Sea proper region due to the presence of the macroalgal species which are well established and adapted in the Baltic Sea despite its low salinity (Kautsky and Kautsky, 2000; Schagerström et al., 2014) (high IA on 11 August 2019 event day). The rocky shorelines of the northern Baltic Sea provides ample habitat for several species of macroalgae, including F. vesiculosus (Kautsky & Kautsky 2000, Torn et al., 2006). Previous studies have documented that certain macroalgae contain high levels of iodine (Ar Gall et al., 2004), of which the kelp *Laminaria digitata* stores the highest amount (Ar Gall et al., 2004; Küpper et al., 1998).

However recent chamber experiments comparing different species of brown algae found that emission rate of I₂ was higher in the case of *F. vesiculosus* when compared to other species like *L. digitata* (Huang et al., 2013). This could possibly explain the high IA concentration recorded by the CI-APiTOF when the air mass was coming from the Northern Baltic Sea region (11 August 2019 and 14 August 2019). High production of macroalgal species is common along the extensive archipelago coastlines of the northern Baltic Sea, and particularly *F. vesiculosus* is likely to contribute with high emission rates, especially when during peak production times when exposed to low sealevels and direct sunlight. However, partitioning the influence of macroalgae and other microalage requires further mechanistic studies. We suggest that marine and coastal regions surrounding the measurement site are capable of producing SA and IA during bloom period, which can initiate NPF.

4 Conclusions

We studied the composition, concentrations and sources of precursor vapors forming aerosols in Helsinki, Finland during the summer of 2019. The source of precursor gases responsible for new particle formations were assessed by analyzing the meteorological parameters, situation of cyanobacterial/algal bloom in the Baltic Sea. Our study recorded several regional, local and burst events and we found that they were connected to elevated concentrations of SA and IA. The burst /spike events occurred simultaneously with high intensity cyanobacterial/algal blooms in the Baltic Sea.

The study draws the following conclusions. 1) Constantly changing algal conditions in Gulf of Bothnia, Gulf of Finland and Baltic Sea could be a significant source for the emission of iodine precursors and DMS. These gases produced by these emissions further oxidize in the atmosphere to form IA and SA, which can be detected by mass spectrometric methods. Interestingly, during marine air mass intrusion with higher residence time over the algal blooms, the gaseous

precursors formed from the biological emissions possibly exceeded the gaseous precursors sourced from anthropogenic emissions at the measurement site. In fact, an overall higher impact of biogenic emissions was noted in this semi-urban site particularly during end of July and mid-August when the bloom intensity decreases and the cyanobacteria/macroalgae start to decay and die (while being exposed to sunlight) and consequently produce more emissions (biogenic SA and IA). 2) Moreover, the meteorological conditions like wind direction and possibly wind speed were identified as the most important parameters influencing the precursor vapor concentration reaching the measurement site and thus determining if NPF occurred. Further we also infer that that the wind direction played an important role in determining the particle concentrations at the study site. Our study reports, that when the air mass travelled over the land with higher residence time of the air mass over the urban areas, it was enriched with SA and organics from proximal-local sources leading to the occurrence of regional and local events (30 June 2019 and 30 July 2019). In contrast, when the air mass travelled over the water bodies, with higher residence times over the cyanobacterial blooms, the air mass was enriched with biogenic IA and/or SA initiating a burst/spike event at the measurement site (11, 14, 15 August 2019). This observation is comparable to other coastal sites like Mace Head, although the NPF events are much stronger in Mace Head, since the measurement site is just at the coast with intensive low tide-high tide periods. 3) The formation rates of 1.5 nm particle and ions suggest that both IA-driven and SA-driven NPF events were neutral nucleation events. 4) The type of phytoplankton species, intensity of the bloom and distance of the bloom from the experimental site is speculated to play an important role in determining the concentrations of precursor gases and thus influence the duration and type of NPF. The IA driven nucleation occurred when the air mass travelled from over the Baltic Sea region, where the coasts are dominated by several species of macroalgae, including F. vesiculosus. The SA rich burst events occurred when air mass travelled over the Gulf of Bothnia which was mainly dominated by the cyanobacteria species Aphanizomenon. 5) Burst/spike events, connected to high IA concentrations, likely led to fast growth of particles potentially to CCN sizes. The role of stabilizing the IA clusters by SA and ammonia in a semi-urban coastal place needs to be further explored. The growth rate of particles was not fully explained by the SA, IA and MSA alone, this applies especially for 3–7 nm or larger particles, indicating that organics might be playing a critical role in the growth of particles in this semi-urban location. We have significantly high ambient concentrations of HOMs in this study, although the detailed descriptions is beyond the scope of this work.

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The role of organics (HOM) in the growth of particles is an active research question. Exploring the sources and characterizing them during a bloom period, when the emission of biogenic volatile organics increase with temperature, is crucial to understand the climate linkages of aerosol

- 794 formation. In order to resolve these links require more quantitative studies are required, which aims
- 795 to understand the correlation between the quality and quantity of cyanobacterial blooms and the
- strength of emissions of aerosol precursors. More systematic studies, partitioning the influence of
- 797 pelagic cyanobacterial blooms and coastal macroalgae on new particle formations, would need to be
- 798 undertaken.
- 799
- 800 Data availability
- 801 Mass spectrometer and air ion spectrometer data related to this article are available upon request to
- 802 the corresponding author. Rest of the data are available for download from
- 803 https://avaa.tdata.fi/web/smart/smear.
- 804 Supplement
- The supplement related to this article is available online at:
- 806 Author contributions
- 807 RCT and TJ, MS designed the experiment, MS, LB, NS, YJT, TC, YJ, JL, ML were involved in the
- 808 instrument installations and performed calibrations, RCT, collected, processed, analyzed and
- interpreted the mass spectrometric data. TC, JS, JL, RCT and ML collected and processed the particle
- data. RCT, LD and KL interpreted the particle data. LD, LB, LQ and XCH preformed the calculations.
- 811 MS, RCT, TJ and MK conceptualized the idea of connecting marine biology and atmospheric
- processes. AN improvised the marine biology section of the paper. CX carried out Flexpart analysis.
- 813 MM contributed to the satellite data procurement and its interpretation. All authors contributed
- 814 commented on the manuscript and improvised the data interpretation.
- 815
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