



Measurement report: Air ion composition during new particle formation events in summertime Cyprus

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Abstract

New particle formation is an important source of atmospheric aerosols, but the chemical factors controlling its frequent occurrence in the Eastern Mediterranean remain poorly constrained. We investigated summertime new particle formation and negative ion composition at the rural Cyprus Atmospheric Observatory site during July–September 2023 using ion and particle size distributions, molecular-level ion composition, trace gases, aerosol properties, and meteorological observations. New particle formation was observed on 19 of 59 measurement days and occurred less frequently in August than in September, consistent with the summertime minimum previously observed in Cyprus. Growth rates increased with particle size, while ion-induced nucleation contributed a median of ~4 % to the total particle formation rate. Events occurred under broadly similar global radiation, sulfur dioxide, ozone, nitrogen oxide, and meteorological conditions, but under lower particulate matter concentrations and condensation sink than non-event days. Molecular-level measurements revealed a pronounced diurnal evolution of atmospheric ion chemistry. Daytime ion composition was dominated by sulfuric acid monomers, dimers, and trimers together with several mixed sulfuric acid–organic clusters. Nighttime spectra were dominated by highly oxygenated organic molecules clustered with nitrate. Positive matrix factorization resolved four factors describing the diurnal changes, including an iodate-containing afternoon transition between sulfuric acid-dominated daytime chemistry and highly oxygenated organic molecule-dominated nighttime chemistry. These measurements provide molecular-level characterization of summertime atmospheric ion chemistry in the Eastern Mediterranean and new insight into the molecular composition of clusters involved in the earliest stages of atmospheric particle formation.



43 **1. Introduction:** New particle formation (NPF) is a fundamental atmospheric process through
44 which nanometer-sized molecular clusters form from gaseous precursors and subsequently
45 grow to climatically relevant sizes (Lee et al., 2019; Spracklen et al., 2008). NPF contributes
46 substantially to global cloud condensation nuclei (CCN) budgets and therefore influences cloud
47 properties, air quality, and climate forcing (Sullivan et al., 2018; Kulmala et al., 2022;
48 Sebastian et al., 2022). It has been estimated that roughly half of global CCN originate from
49 atmospheric nucleation processes, highlighting the importance of understanding the chemical
50 pathways and environmental conditions that govern cluster formation and early growth
51 (Gordon et al., 2017; Westervelt et al., 2014). Despite major advances in instrumentation and
52 theory, the molecular-level mechanisms of NPF remain incompletely resolved across diverse
53 atmospheric environments, particularly in warm regions outside the boreal zone.

54
55 In the Eastern Mediterranean and Middle East (EMME) region, NPF is a frequent phenomenon
56 influenced by complex air mass origins and strong photochemistry (Kalkavouras et al., 2021;
57 Baalbaki et al., 2021; Pikridas et al., 2012; Kalivitis et al., 2015; Deot et al., 2025; Ajith et al.,
58 2024). Long-term measurements in Cyprus revealed that NPF occurs throughout the year, with
59 the highest NPF frequency in spring and lowest in summer (Baalbaki et al., 2021; Deot et al.,
60 2025). However, to our knowledge, direct molecular-level measurements of the vapours and
61 initial clusters involved in atmospheric nucleation have not previously been reported from the
62 EMME area. This lack of chemical information limits the identification of the mechanisms
63 responsible for particle formation in the region and complicates their representation in
64 atmospheric models. Comprehensive characterization of precursor vapours and cluster
65 composition is therefore particularly needed during summer, when intense solar radiation
66 enhances atmospheric oxidation chemistry, yet NPF occurs less frequently (Baalbaki et al.,
67 2021)

68
69 Sulfuric acid (SA) is a key precursor in atmospheric nucleation, and field and laboratory studies
70 have consistently shown that particle formation rates correlate with neutral SA concentrations
71 (Jokinen et al., 2017; Kulmala et al., 2013). However, detecting the molecular clusters involved
72 in the earliest stages of atmospheric particle formation remains challenging because of their
73 extremely low concentrations and short lifetimes. Much of the molecular-level understanding
74 of atmospheric nucleation mechanisms has therefore been derived from measurements of
75 naturally charged clusters using the Atmospheric Pressure interface Time-of-Flight mass
76 spectrometer (APi-TOF (Junninen et al., 2010). Owing to its high sensitivity to ambient ions
77 and ion clusters, the APi-TOF can detect initial molecular clusters that are often below the
78 detection limit of chemical-ionization instruments used to measure neutral vapours and
79 clusters. These ion measurements have demonstrated that the formation and stability of sulfuric
80 acid clusters depend not only on SA availability but also on temperature, relative humidity, ion
81 concentrations, and the presence of stabilizing compounds (Duplissy et al., 2016; Tuovinen et
82 al., 2024; Jokinen et al., 2018; Kirkby et al., 2016). In particular, basic vapours such as
83 ammonia (NH₃) and amines can stabilize sulfuric acid clusters by reducing their evaporation
84 rates and thereby enhance nucleation (Almeida et al., 2013; Kürten et al., 2014). Highly
85 oxygenated organic molecules (HOMs), formed through the oxidation of volatile organic
86 compounds by ozone, hydroxyl radicals, and nitrate radicals, may also participate in nucleation
87 and early particle growth, either independently or together with SA (Kirkby et al., 2016; Tröstl
88 et al., 2016; Lehtipalo et al., 2018). Parameterization efforts across multiple global
89 environments demonstrate that SA remains a primary driver, but that meteorology and
90 additional vapours modulate formation rates substantially (Li et al., 2025). These findings
91 highlight the importance of knowing the composition of both neutral and charged clusters to
92 fully understand atmospheric nucleation mechanisms.



93 Despite high solar radiation and elevated SA (proxy) concentrations in summer in Cyprus, the
94 lower NPF frequency points towards additional precursor vapor or air mass transport
95 controlling NPF in this region (Baalbaki et al., 2021). In this report, we study nanoparticles and
96 naturally charged ions measured from a rural background site of the Cyprus Atmospheric
97 Observatory in Agia Marina Xyliatou (CAO-AMX), carried out during the summer and autumn
98 of 2023. We focus on the combination of ion number size distribution data with molecular-
99 level chemical composition and binned positive matrix factorization (PMF) analysis in order
100 to shed light into the characteristics of NPF events at the site and aim to identify the
101 composition of cluster forming ions during these events.

102

103 2. Materials and methods

104 **2.1 Measurement site:** Measurements were conducted at the CAO-AMX station between 27
105 July 2023 – 29 September 2023. CAO-AMX (35.038692 °N, 33.057850 °E, 532 m above mean
106 sea level) is one of the four stations under CAO that provide continuous measurements of trace
107 gases, and aerosol physical, chemical and optical properties following the Aerosol, Clouds, and
108 Trace Gases Research Infrastructure (ACTRIS) (Laj et al., 2024) and Global Atmosphere
109 Watch (GAW, Schultz et al., 2015). CAO-AMX is a rural background site with minimal
110 influence of local pollution sources but strongly affected by the regional and long-range
111 transport. More details on the CAO-AMX measurement site and instrument installations can
112 be found in Deot et al. (2025). Note that all observations from CAO-AMX are recorded in
113 Coordinated Universal Time (UTC) and all the data presented in this article is thus UTC, not
114 local time (Easter European Summer Time = UTC+3).

115

116 **2.2 Atmospheric Pressure interface Time-Of-Flight (APi-TOF) mass spectrometer:** APi-
117 TOF mass spectrometer (“LTOF”, ToFwerk, Switzerland) is a high-resolution analytical
118 instrument developed to identify ambient atmospheric ions and naturally occurring ion clusters
119 with high mass accuracy and resolution (Junninen et al., 2010). The ambient ion mode allows
120 the detection of ambient anions and clusters without artificial ionization, providing a
121 representation of naturally occurring atmospheric charging where the lowest proton affinity
122 compounds dominate the spectra (e.g., strong acids).

123

124 The APi samples the atmospheric air at a rate of 0.8 liters per minute (lpm) through the critical
125 orifice (0.3 mm diameter). The sample air flow rate to the critical orifice was kept at 20 lpm.
126 Ions are transferred from passing the APi through in three different stages that progressively
127 reduce the pressure before entering the high vacuum towards the TOF chamber, which is
128 maintained at a pressure of around 10^{-6} mbar to prevent ion collisions with gas molecules and
129 unobstructed ion flight. In the TOF chamber, ions are accelerated and separated according to
130 their mass-to-charge ratio (m/z) with high mass resolution (~ 8500 Th/Th). M/z are reported in
131 Thomson (Th) where 1 Th corresponds to 1 Dalton per elementary charge ($1 \text{ Da } e^{-1}$).

132

133 The APi-TOF data was analysed using the MATLAB tofTools (Junninen et al., 2010) and
134 IGOR Pro Tofware software (version 4.0.3; Stark et al., 2015). In tofTools, data were averaged
135 to 60 min before mass calibration. Mass calibration was performed using an internal calibration
136 approach based on the following, known ions: NO_2^- , NO_3^- , $(\text{HNO}_3)\text{NO}_3^-$, HSO_4^- , and
137 $(\text{H}_2\text{SO}_4)\text{HSO}_4^-$, resulting in a mass accuracy better than ± 8 ppm (parts per million) across the
138 analysed mass range. We further applied a binned positive matrix factorization (PMF) method
139 to the mass spec data following Katz et al. (2023) and Zhang et al. (2019). Binning of the data
140 has been performed in Tofware using the binDatEx workflow after performing mass calibration
141 with a bin size of 0.01 m/z prior to PMF analysis. In this approach, the PMF model is performed
142 on binned data directly, without requiring any prior chemical information or peak fitting.



2.3 Neutral cluster and Air Ion Spectrometer (NAIS): Ion and neutral particle size distributions were measured using a NAIS (Airel Ltd., Estonia). The NAIS measures ions and naturally charged particles in the diameter range of 0.8 – 42 nm (109 size bins obtained from 25 electrometers) (Manninen et al., 2016; Mirme and Mirme, 2013). Ambient air was sampled through a 30 mm diameter and 80 cm long sampling inlet tube. The system operated at a volumetric flow rate of 54 lpm. Concentrations of particles with sizes smaller than ~2 nm were excluded due to known artefacts from the corona charger. Raw counts were inverted using the NAIS SPECTOPS algorithm and corrected for inlet losses (Gormley and Kennedy, 1948).

2.4 Scanning Mobility Particle Sizer (SMPS): The SMPS system consists of a TSI 3083 long differential mobility analyzer (DMA) coupled with a TSI 3789 condensation particle counter (CPC). It measures particle size distributions in the diameter range of 10- 778 nm (61 size bins). The aerosol and sheath flow rates were checked weekly and maintained at 1.0 and 4.8 lpm, respectively. Ambient air was sampled through a ~3 m inlet tube and drying was achieved using a Nafion dryer (RH < 40%), following ACTRIS standard procedures for in-situ aerosol sampling and analysis.

2.5 Ancillary measurements: Collocated trace gas measurements (within a 50 m radius), including sulfur dioxide (SO₂), carbon monoxide (CO), ozone (O₃), oxides of nitrogen (NO_x = NO+NO₂), particulate matter of aerodynamic diameter less than 2.5 µm and 10 µm (PM_{2.5} and PM₁₀) as well as meteorological parameters (temperature, relative humidity, solar radiation, precipitation, wind speed, and wind direction), were obtained from the national air quality monitoring network operated by the Department of Labour Inspection (DLI) of Cyprus. Details of trace gas and meteorological parameters measurement techniques and procedures are given in (Vrekoussis et al., 2022).

2.6 Event type identification: The different event types were identified by visual inspection of particle size distribution contour plots (Dal Maso et al., 2005), and classified as NPF event, non-event, and undefined days. A day was identified as an NPF event day when a rapid increase in particle number concentrations was observed in the nucleation mode (<20 nm), followed by the continuous growth of these particles to larger sizes. A day without a significant increase in nucleation mode particles was identified as a non-event day. A day that could not be classified into either of these two types was classified as an undefined day.

2.7 Ion and particle growth rate (GR) and formation rate (J): The particle GR of newly formed ions or particles is determined from the measured ion or particle size distributions (Hirsikko et al., 2007; Kulmala et al., 2012). Particle GRs were obtained by a linear least-squares fit through the temporal evolution of particle mode diameter of the measured particle size distribution. Ion GRs, particularly in the sub-3-nm range, were determined by the maximum-concentration method (Kulmala et al., 2012; Lehtinen and Kulmala, 2003). Furthermore, we have calculated particle GR in different size ranges (2-3 nm, 3-7 nm, and 7-20 nm). The particle formation rate at 2 nm (J₂) was calculated by taking into account the particle number concentrations in the size range 2-3 nm, coagulation scavenging to the large particles, and the growth rate (Kulmala et al., 2007) given by the following equation;

$$J_2 = \frac{dN_{2-3}}{dt} + CoagS_2 \cdot N_{2-3} + \frac{f}{1nm} GR_{2-3} \cdot N_{2-3} \quad (1)$$

where N₂₋₃ is the number concentration of 2-3 nm particles, CoagS₂ is the coagulation sink of 2 nm particles (Kulmala et al., 2001), and GR₂₋₃ is the particle growth rate in the 2-3 nm size



range, all these were obtained from the NAIS measurements. The factor f indicates the fraction of 2-3 nm particles that were activated for the growth, assumed to be unity in this work.

For charged particles, both ion-ion recombination and the charging of the neutral particles in the 2-3 nm size range need to be considered (Manninen et al., 2009). The formation rate of 2-3 nm charged particles can be expressed as,

$$J_2^\pm = \frac{dN_{2-3}^\pm}{dt} + CoagS_2 \cdot N_{2-3}^\pm + \frac{f}{1nm} GR_{2-3} \cdot N_{2-3}^\pm + \alpha \cdot N_{2-3}^\pm \cdot N_{<3}^\mp - \beta \cdot N_{2-3} \cdot N_{<2}^\pm \quad (2)$$

where the superscript $\pm$ indicates to positively and negatively charged particles, respectively, $N_{2-3}^\pm$ is the 2-3 nm ion concentrations, $N_{<2}^\mp$ and $N_{<3}^\mp$ is the ion concentrations below 2 nm and 3 nm, respectively. α and β are the ion-ion recombination coefficient and the ion-neutral attachment coefficient, assumed as $1.6 \times 10^{-6} \text{ cm}^3 \text{ s}^{-1}$ and $0.01 \times 10^{-6} \text{ cm}^3 \text{ s}^{-1}$, respectively (Tamm et al., 2005).

2.8 Coagulation sink (CoagS) and condensation sink (CS): The CoagS describes the rate at which nanometer-sized aerosol particles are removed through coagulation with pre-existing particles and it was determined by the following equation (Kulmala et al., 2001).

$$CoagS_d = \sum_{d_i}^{d_j} K(d_i, d_j) \cdot N_j \quad (3)$$

where $K(d_i, d_j)$ is the coagulation coefficient (kernel) of particles with sizes d_i and d_j , calculated by using the formula of Fuchs (1964).

The CS quantifies the ability of the existing aerosol population to remove condensable vapors from the atmosphere through condensation onto particle surfaces and it was determined by the following equation (Kulmala et al., 2001),

$$CS = 2\pi D \cdot \sum_{d_j} \beta_{m,d_i} \cdot d_j \cdot N_{d_j} \quad (4)$$

where D is the diffusion coefficient of the condensing vapour, assumed to be H_2SO_4 , and $0.104 \text{ cm}^2 \text{ s}^{-1}$ and β_m represents a transition regime correction calculated by

$$\beta_m = \frac{1+K_n}{1+1.677 \cdot K_n + 1.333 \cdot K_n^2} \quad (5)$$

where K_n is the Knudsen number given by $K_n = 2\lambda/d$, λ is mean free path (Seinfeld and Pandis, 2006).

2.9 Contribution of ion-induced nucleation (IIN): The total nucleation rate can be expressed as the sum of neutral and ion-induced nucleation rates (Manninen et al., 2009). Therefore, the contribution of ions to particle formation can be expressed as the sum of the formation of charged ions to the total formation rate (J_2).

$$IIN = \frac{J_2^+ + J_2^-}{J_2} \quad (6)$$



3. Results and Discussion

3.1. New particle formation events: Ion and particle number size distributions measured by NAIS and SMPS were used to identify NPF events (Fig. 1). From 59 valid observation days, we classified 19 days (32%) as NPF, 24 days (41%) as non-events, and 16 days (27%) as undefined. The monthly fractional occurrence of the different event types is presented in Fig. 2. NPF events were observed less frequently in August (summer) than in September (autumn), consistent with previous multi-year observations from the same site (Baalbaki et al., 2021). Several studies have shown that the occurrence of NPF often peaks during spring and autumn, when the balance between precursor availability and particle sink is more favorable for nucleation, and the lowest during the summer and winter (Kanawade et al., 2012; Kerminen et al., 2018; Nieminen et al., 2018; Lee et al., 2019), with some exceptions showing the highest frequency in summer (Tang et al., 2023).

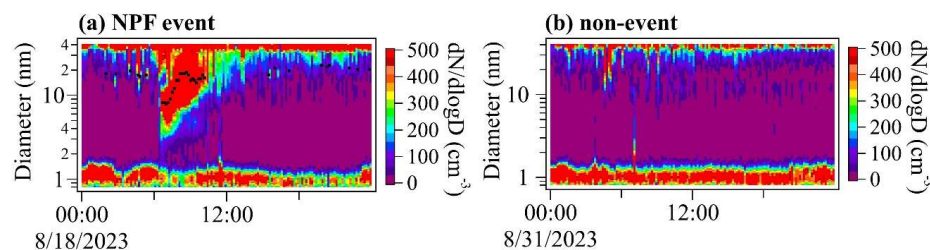


Figure 1. Typical event and non-event day. Examples of typical (a) NPF event (18 August) and (b) non-event (31 August) in UTC time, showing ion number size distributions. Black dots in (a) show ion mode diameter.

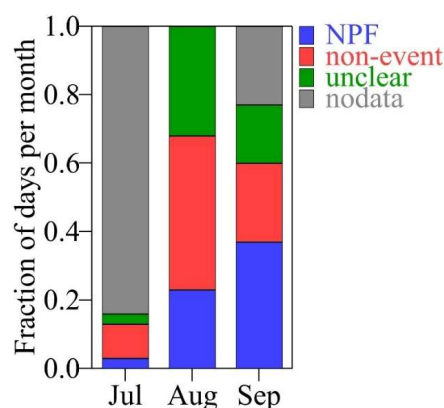


Figure 2. Monthly event classification. Stacked-bar plot of the fraction of days per month of different event types.

3.2 Observed growth rates and formation rates: We present the size-dependent negative ion and particle GRs in Fig. 3 that are key parameters used to characterize NPF events. The GR increases with particle size, with median values ranging between 1.2 - 3.1 nm h⁻¹ and 0.6 - 1.7 nm h⁻¹ for negative ions and particles, respectively. The total particle formation rate (J_2) on the observed NPF event days ranged from 3.3×10^{-3} - $12.3 \text{ cm}^{-3} \text{ s}^{-1}$, while negative (J_2, neg) and positive (J_2, pos) ion formation rates ranged from 1.7×10^{-5} - $0.17 \text{ cm}^{-3} \text{ s}^{-1}$ and 3.6×10^{-5} - $0.12 \text{ cm}^{-3} \text{ s}^{-1}$, respectively. This results in an IIN contribution ranging from 0.03 to 0.12, with a median value of 0.04 (4%). The median charged contribution to particle formation at 2 nm was



therefore approximately 4%, indicating that charged particles constitute only a minor fraction of the particle formation flux at this size. However, this estimate does not uniquely constrain the initial nucleation mechanism, because ion-ion recombination may neutralize clusters before they reach 2 nm. Similar low charged contributions at 2 nm have been reported in previous field measurements, with the contribution of ion-induced nucleation to the total particle rate of 2 nm typically ranging between 1 and 30% (Manninen et al., 2010; Victor et al., 2024; Manninen et al., 2009; Iida et al., 2006). This suggests that while ions may initiate or enhance early cluster formation, the dominant NPF pathway appears to be neutral nucleation.

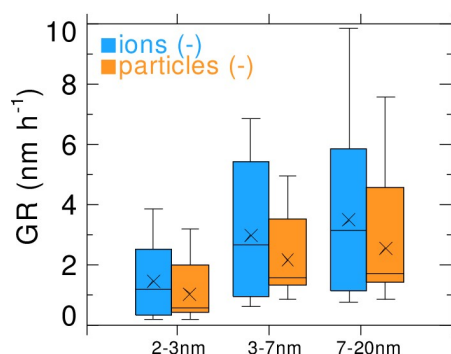


Figure 3. Size-dependent growth rates. Size-dependent ion and particle growth rates during observed NPF events for negative polarity measurements. The solid horizontal line indicates the median, the cross sign indicates the mean, the box indicates the 25th and 75th percentiles, the whiskers indicate the 10th and 90th percentiles.

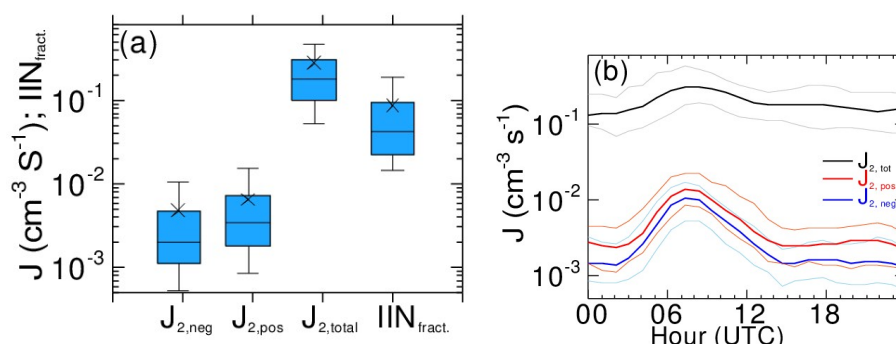


Figure 4. Formation rates. (a) Box-whisker plot of negative and positive ions and total particle formation rates at 2 nm, and the fractional contribution of ion-induced nucleation during observed NPF events. The solid horizontal line indicates the median, the cross sign indicates the mean, the box indicates the 25th and 75th percentiles, and the whiskers indicate the 10th and 90th percentiles. (b) Median (thicker lines) diurnal variation of negative and positive ion and total particle formation rates at 2 nm. Thin lines represent 25th and 75th percentiles.

3.3 Overview of trace gases, dust and meteorological conditions: Here, we provide an overview of the trace gases, PM concentrations and meteorological variables to characterize the atmospheric conditions under which summertime NPF was observed. The temporal evolution of ion number size distributions, ion and aerosol number concentrations, aerosol sinks (CoagS and CS), trace gas concentrations, PM_{2.5} and PM₁₀, and meteorological conditions during the measurement campaign is summarized in Fig. 5.



296

297 The campaign period represents typical summer conditions in Cyprus that is characterized by
298 strong solar radiation (Fig. 5i), very high daytime temperatures (Fig. 5h), and generally low
299 background aerosol concentrations with mode diameter approximately 100 nm (Fig. 5c). It is
300 notable that NPF events were observed during the summer season at daytime temperatures
301 exceeding 30°C (Fig. 5h). Similar observations have been reported in other Mediterranean
302 environments. Across several sites in Greece, summertime NPF occurred during measurement
303 periods characterized by temperatures reaching all the way up to 40°C, and temperature
304 generally did not differ significantly between event and non-event days (Aktypis et al., 2024).
305 NPF has also been observed in Barcelona at an average event temperature of approximately
306 303 K (~30°C, Brean et al., 2020). These observations demonstrate that high ambient
307 temperatures do not necessarily prevent NPF under Mediterranean conditions, although
308 elevated temperatures are expected to enhance cluster evaporation. Overall, meteorological
309 parameters show relatively stable conditions, with an occasional rainfall episode causing short-
310 term decreases in PM, aerosol sink, and trace gas levels. In addition, episodes of high dust
311 loading, elevated and at ground level, were observed during September.

312

313 Trace gas concentrations displayed episodic variability (Fig. 5e-f), suggesting fluctuations in
314 precursor availability for nucleation and early particle growth. PM concentrations and aerosol
315 sinks were relatively higher during summer than in early autumn (Fig. 5g). Both the
316 condensation sink (CS) in Cyprus ($\sim 1 \times 10^{-2} \text{ s}^{-1}$) and the $\text{PM}_{2.5}$ are approximately 2–5 times
317 higher than typical values observed in boreal environments such as Hyytiälä, Finland (Dal
318 Maso et al., 2005; Kulmala et al., 2025), and CS comparable to other EMME sites such as
319 Amman, Jordan (Hussein et al., 2020) or Thessaloniki and Ioannina (Aktypis et al., 2024).
320 Compared to the cleaner and more studied boreal environments, Cyprus exhibits substantially
321 higher SO_2 (~0.5–1 ppbV) and NO_x (~2 ppbV) concentrations, together with elevated ozone
322 levels (30–70 ppbV), indicating enhanced photochemical activity and a greater potential for
323 SA and secondary organic aerosol production despite a higher CS. Our observations are
324 consistent with previous findings that regional NPF events often occur under conditions of
325 strong photochemical activity and low pre-existing particle surface area (Dal Maso et al., 2005;
326 Kulmala et al., 2012).

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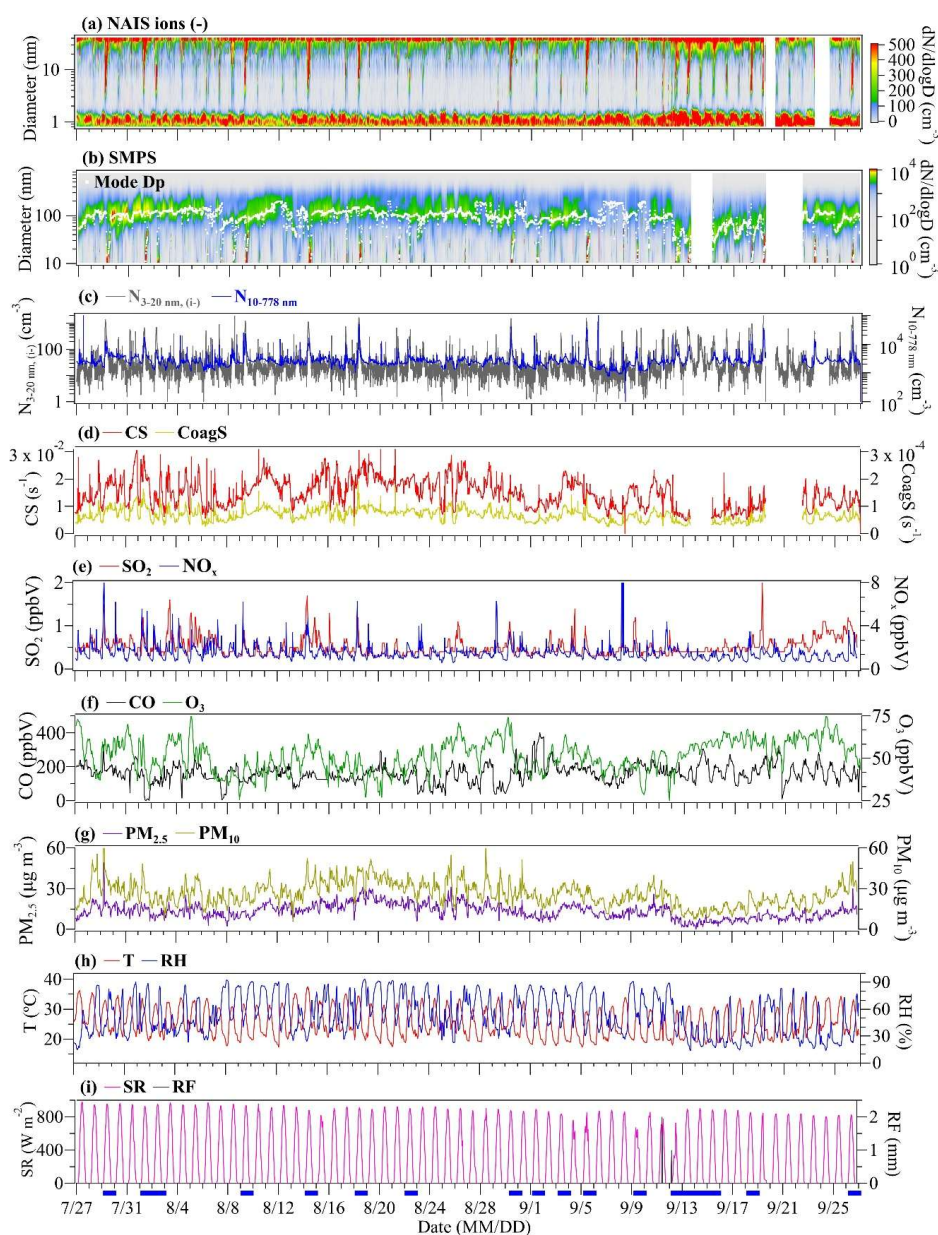


Figure 5. Time evolution of (a) negative polarity ions number size distributions, (b) particle number size distributions from the SMPS (c) number concentrations of negative ions in the size range of 3-20 nm ($N_{3-20 \text{ nm}, (i-)}$) and aerosol particles in the size range of 10-778 nm ($N_{10-778 \text{ nm}}$), (d) condensation sink (CS) and coagulation sink (CoagS), (e) sulfur dioxide (SO_2) and oxides of nitrogen (NO_x), (f) carbon monoxide (CO) and ozone (O_3), (g) particulate matter mass concentrations less than aerodynamic diameter 2.5 μm ($\text{PM}_{2.5}$) and 10 μm (PM_{10}), (h) temperature (T) and relative humidity (RH), and (i) solar radiation (SR) and rainfall (RF) during the summer of 2023. The horizontal blue lines indicate the observed NPF events.



4. Comparison of NPF event and non-event conditions

4.1. Ion number concentrations: Across size ranges over 2 nm, ion concentrations increase sharply during NPF events compared with non-event days, however, ion concentrations below 2 nm are comparable between the events (Fig. 6). A pronounced morning maximum occurs between 6:00 and 10:00 UTC, particularly for the intermediate ions (2–3 and 3–7 nm), indicating the onset of particle formation. The shift in peak ion concentration to larger size indicates subsequent growth of freshly formed sub-3nm ions to larger sizes. In contrast, non-event days show lower concentrations and much weaker diurnal variability. This indicates cluster formation and growth were suppressed, likely due to unfavorable conditions such as a higher CoagS or limited availability of precursor vapors. These observations agree with previous studies showing that NPF events are characterized by elevated concentrations of ions, and those coincide with enhanced production of sulfuric acid and oxidized organic vapors that drive particle formation and early growth (Ehn et al., 2014; Riccobono et al., 2014; Almeida et al., 2013).

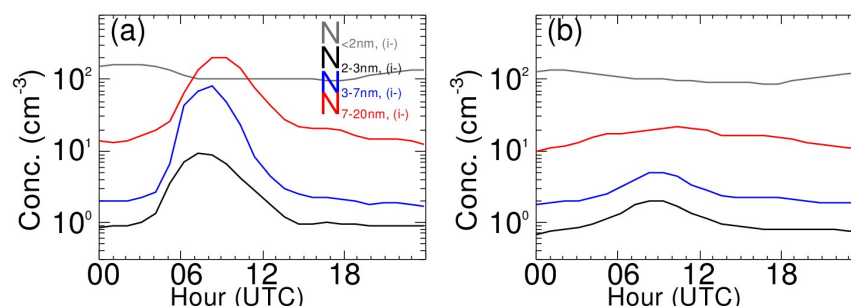


Figure 6. Median diurnal variation of size-segregated anion number concentrations during observed (a) NPF events and (b) non-events.

4.2 Trace gases, meteorological conditions, and particulate matter: NPF is strongly dependent on meteorological conditions (Kanawade et al., 2012; Kulmala et al., 2004; Stanier et al., 2004). Solar radiation and photochemistry have been identified as critical factors in the NPF processes (Jokinen et al., 2017; Kulmala et al., 2004; Nieminen et al., 2018). Here, we present the median diurnal variation of trace gases, meteorological parameters, and PM during NPF event and non-event days during summer, as shown in Fig. 7.

Our results show that NPF events occur under cleaner conditions with lower PM concentrations (Fig. 7g), and higher SO₂ and NO_x concentrations (Fig. 7a–b) which is consistent with field observations in continental environments (Kulmala et al., 2004). A small increase in SO₂ can significantly enhance nucleation rates (Sipilä et al., 2010), while the higher NO_x concentrations can favor oxidation chemistry that produces highly oxygenated organic molecules, which can contribute to particle growth (Yan et al., 2021). Similar O₃ levels in the morning hours (Fig. 7c) and high daytime temperature (Fig. 7d) further demonstrate strong photochemical activity at the site (regardless of the event type), which is typical during summer. The lower relative humidity also favors NPF at our site.

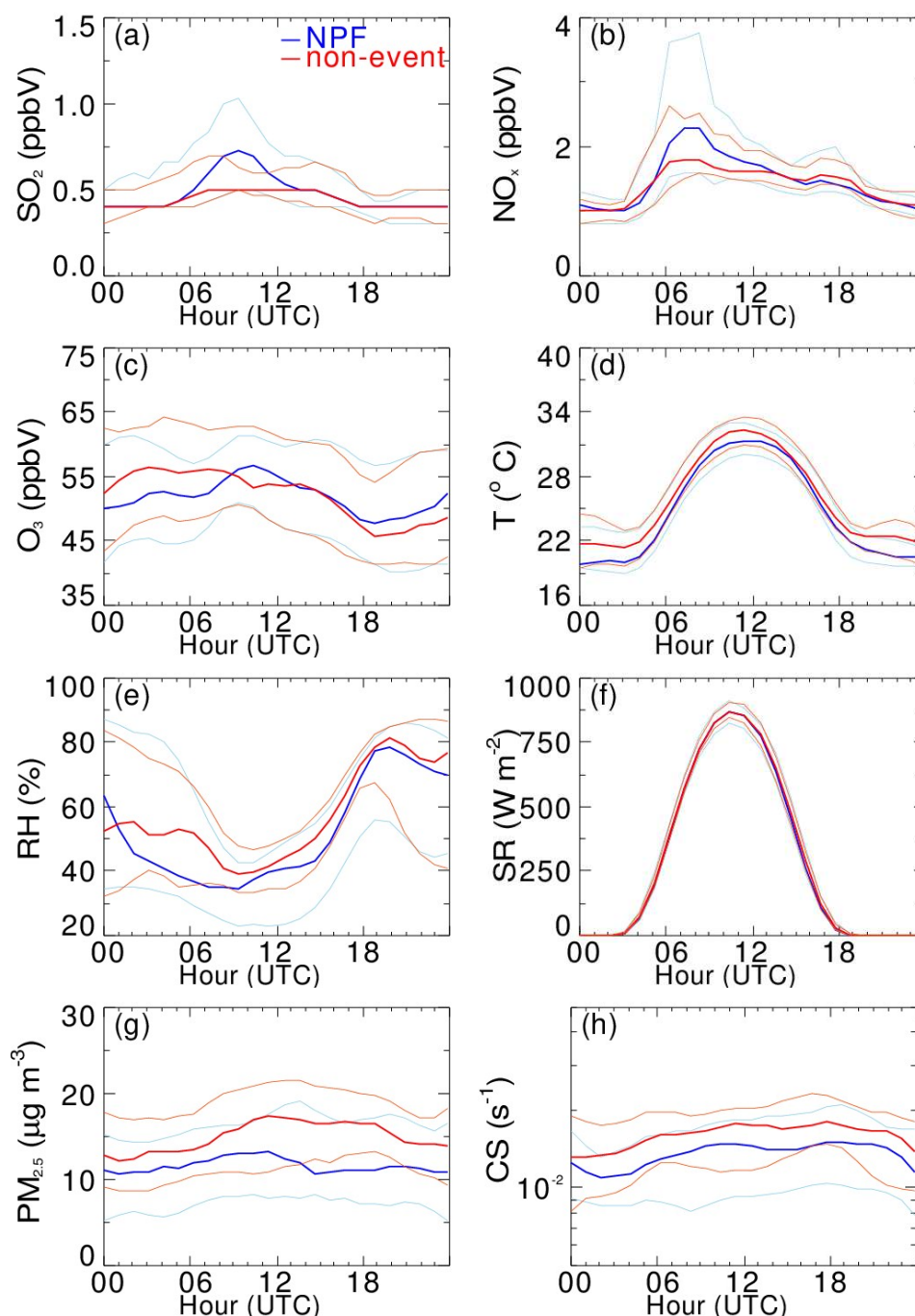


Figure 7. Median (thick lines) diurnal variation of trace gases, meteorological parameters and particulate matter for observed NPF events (blue lines) and non-events (red lines); ((a) sulfur dioxide (SO_2), (b) oxides of nitrogen (NO_x), (c) ozone (O_3), (d) temperature (T), (e) relative



humidity (RH), (f) solar radiation (SR), (g) particulate matter of aerodynamic diameter less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$), and SMPS derived condensation sink (CS). Thin lines represent 25th and 75th percentiles.

In addition, we studies wind-roses comparing NPF and non-event days shows a similar wind direction, but a slightly lower wind speed during NPF days (Fig. 8). In summary, the median diurnal variation of different meteorological parameters does not reveal a significant difference between NPF event and non-event days, showing that atmospheric conditions may not be controlling factors for NPF occurrence in summer at our site.

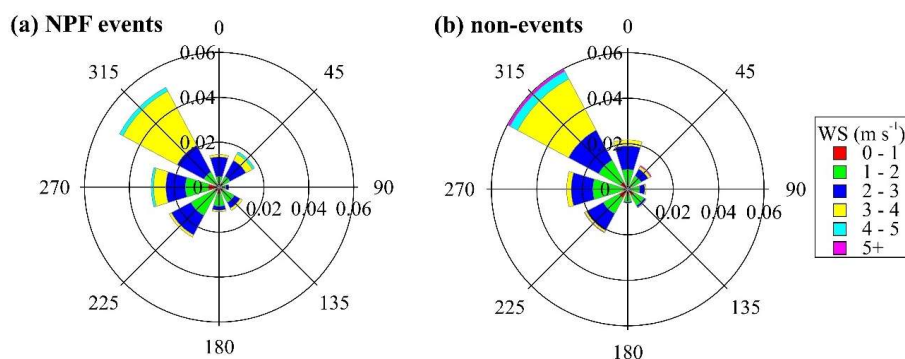
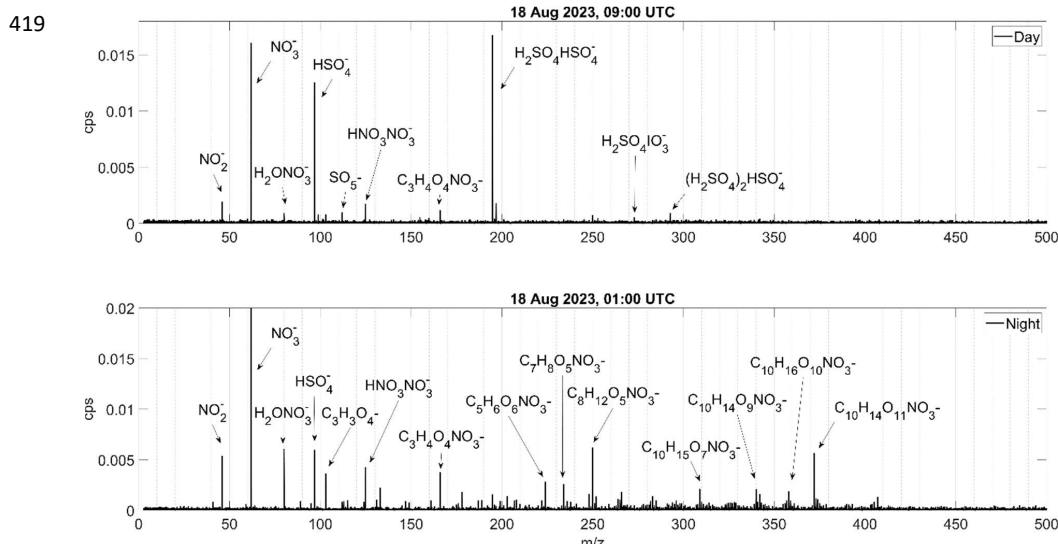


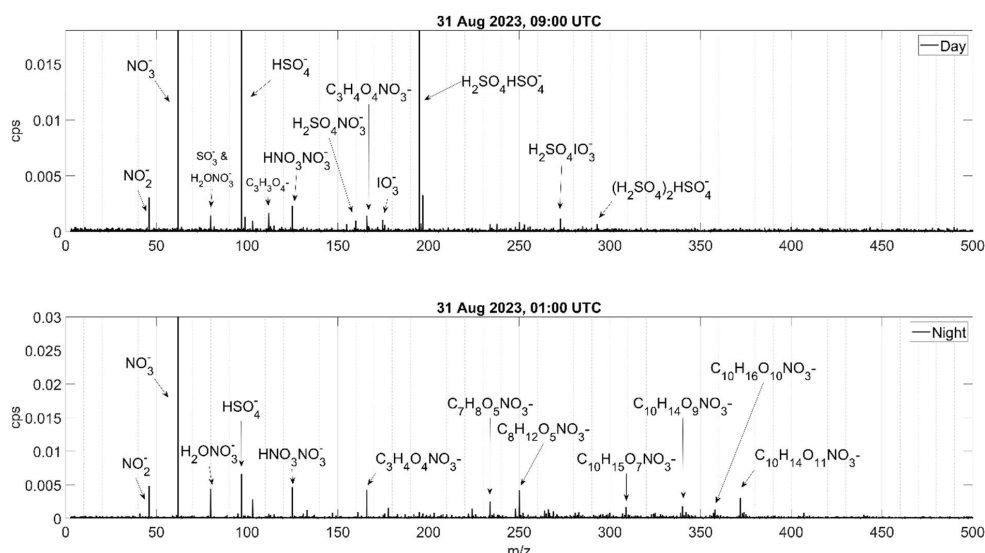
Figure 8. Wind roses. Wind-rose plot showing wind direction (degrees) and speed (WS, m s^{-1}) for NPF events and non-events.

4.3 Chemical composition of anions: Figures 9 and 10 present typical mass spectra during NPF and non-event days presented in Fig. 1. In addition, spectra from the preceding nights are included to provide a broader view of the ion composition, because naturally charged ion spectra reflect the atmospheric charge distribution. This means that the strongest acids dominate the daytime signal, while many less acidic compounds, including most HOMs, remain largely obscured during the day.

Both NPF and non-event daytime spectra are relatively clean and very similar in composition, with charge concentrated in only a few inorganic acid-related ions. The three dominant daytime peaks (NO_3^- , HSO_4^- and $\text{H}_2\text{SO}_4\text{HSO}_4^-$) showed nearly identical relative intensities during the NPF event and non-event day, although their absolute signal levels were consistently higher under non-event conditions when the total ion count is higher. The ion composition is overall dominated by inorganic species, particularly bisulfate (HSO_4^-) and nitrate (NO_3^-) ions and their clusters during the example days. It is notable that we do not detect higher multimer signals after sulfuric acid trimer ($(\text{H}_2\text{SO}_4)_2\text{HSO}_4^-$), and that the sulfuric acid tetramer — ammonia cluster ($\text{NH}_3(\text{H}_2\text{SO}_4)_3\text{HSO}_4^-$) is only shortly detectable during four days out of the 19 NPF events. No higher molecular weight sulfuric acid – ammonia (or any amine) clusters were detected during the events this summer. In addition, we observe some smaller contributions from low molecular weight organic acids (e.g., malonate ion) and their clusters with nitrate ions. A small contribution of iodate (IO_3^-) and its cluster with sulfuric acid is also observed, but more during the non-event days. No significant HOM signals were detected over the two-hour averaging period on either NPF or non-NPF day time spectra. As expected, HOMs are clearly detectable in the night time spectra.



420
 421 **Figure 9. NPF day and night spectra.** Example spectra of an NPF day and the night before
 422 on 18 Aug 2023. During the day, ions mostly consist of bisulfate and nitrate ions and their
 423 multimers with traces of iodine and malonic acid (top panel). In the night, the signal is
 424 distributed to more compounds, including a multitude of HOM alongside abundant nitrate
 425 peaks. The data presents a 120 min average, and the night spectrum has been zoomed in on the
 426 y axis to better show organic compounds.



427
 428 **Figure 10. Non-event day and night spectra.** Example spectra of a non-event day and the
 429 night before on 31 Aug 2023. During the day, ions mostly consist of bisulfate and nitrate ions
 430 and their multimers with traces of iodine and malonic acid (top panel). In the night, the signal
 431 is distributed to more compounds, including a multitude of HOM alongside abundant nitrate
 432 peaks. The data presents a 120 min average, and the night spectrum has been zoomed in on the
 433 y axis to better show organic compounds.



In contrast, the night time spectra prior to the NPF or non-event days exhibit some differences in ion composition. A large number of HOMs appear during the night, together with enhanced nitrate ion signals. Prior to NPF event days, the ion signal is distributed across a wider range of organic compounds than prior to a non-event day.

4.4 Positive matrix factorization (PMF): The example mass spectra (Figure 9 & 10) provide a direct view of the daytime spectra that strongly dominated by a few abundant inorganic ions. Consequently, weaker ions and clusters that show systematic temporal patterns during NPF events are difficult to distinguish from individual spectra alone. To resolve these recurring chemical patterns, PMF was applied to the binned, time-resolved API-TOF spectra from all NPF event days. The analysis separated four factors representing distinct chemical regimes in the diurnal evolution of the ion population (Fig. 11). The suggested fingerprint ions for each factor are listed in Table 1.

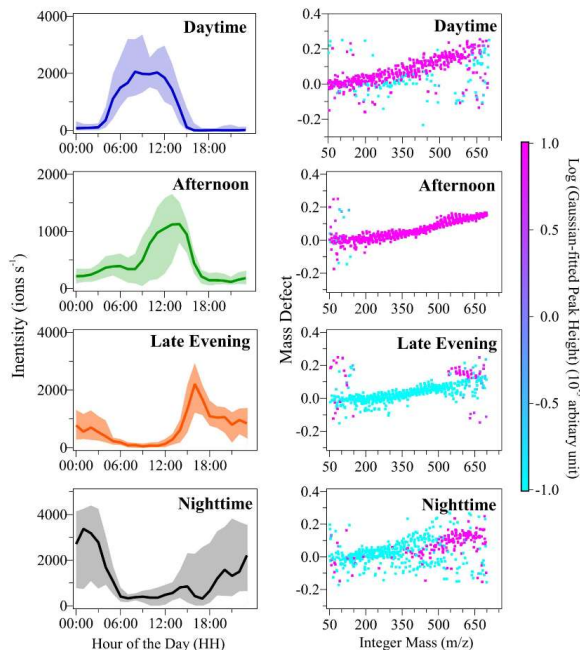
The daytime factor, corresponding to the period when NPF events consistently started, is dominated by sulfuric acid and its molecular clusters, including HSO_4^- , $(\text{H}_2\text{SO}_4)\text{HSO}_4^-$ and $(\text{H}_2\text{SO}_4)_2\text{HSO}_4^-$. Several mixed sulfuric acid–organic clusters, such as $(\text{C}_2\text{H}_2\text{O}_2)\text{HSO}_4^-$, $(\text{C}_3\text{H}_4\text{O}_4)\text{HSO}_4^-$ and $(\text{C}_5\text{H}_6\text{O}_6)\text{HSO}_4^-$, were also identified and classified in the daytime factor. These observations provide direct molecular evidence that sulfuric acid chemistry, together with small organic compounds, dominates the earliest stages of particle formation under summertime conditions in Cyprus. While sulfuric acid monomers and dimers have been observed previously in field measurements (Jokinen et al., 2018; Junninen et al., 2010), direct ambient observations of both sulfuric acid multimers and mixed sulfuric acid–organic clusters remain relatively rare (Bianchi et al., 2016, 2017; Schobesberger et al., 2013). The mixed clusters likely represent sulfuric acid associated with small oxygenated organic compounds, for example, the composition $\text{C}_2\text{H}_2\text{O}_2$ is consistent with glyoxal, suggesting that sulfuric acid may interact with small carbonyl compounds already during the earliest stages of atmospheric cluster formation. Such interactions have been proposed to enhance molecular complexity during nucleation and early particle growth (Elm et al., 2020).

Despite the abundance of sulfuric acid clusters, no sulfuric acid clusters containing ammonia or amines were detected in the PMF results. This was expected from the individual event analysis, where only four events had minor peaks at $\text{NH}_3(\text{H}_2\text{SO}_4)_3\text{HSO}_4^-$ mass. Consequently, the ion composition suggests that sulfuric acid-driven cluster formation proceeds largely without detectable stabilization by organic compounds rather than the common base compounds under the investigated conditions.

The afternoon factor marks a transition from sulfuric acid-dominated chemistry towards different clustering. Besides sulfuric acid-related ions, this factor contains iodate (IO_3^-) together with mixed iodate clusters ($(\text{HIO}_3)\text{NO}_3^-$ and $(\text{HIO}_3)\text{HSO}_4^-$), pointing towards an increasing contribution of iodine chemistry later in the day. In contrast, the nighttime factor is characterized by HOMs clustered with nitrate ions, including several large C_{10} HOM monomers, likely originating from monoterpene oxidation. The dominance of nitrate clustered HOMs demonstrates a pronounced shift from daytime sulfuric acid chemistry to nighttime organic oxidation chemistry. The late evening factor represents an intermediate stage between these regimes, containing both small oxygenated organic ions and the first appearance of larger HOM-derived nitrate clusters. Together, the four PMF factors reveal a clear diurnal evolution of atmospheric ion chemistry, from sulfuric acid-driven cluster formation during daytime NPF to HOM-dominated oxidation chemistry after sunset.



484



485

486 **Figure 11. Binned positive matrix factorization of all NPF event days.** The left panels show
487 the median diurnal intensity profiles (solid lines) of the four PMF factors, with shaded areas
488 indicating the interquartile range (25th–75th percentile). The right panels present the
489 corresponding factor profiles as mass defect versus integer mass (m/z), with colors indicating
490 the logarithm of the Gaussian-fitted peak height.

491 **Table 1. Suggested fingerprint ions for the four PMF factors.** Ions marked with an asterisk
492 (*) are shared between more than one PMF factor.

493

Daytime Factor	Afternoon Factor	Late evening Factor	Nighttime Factor
HSO ₄ ⁻ (C ₂ H ₂ O ₂)HSO ₄ ⁻ (HNO ₃)HSO ₄ ⁻ (H ₂ SO ₄)HSO ₄ ⁻ (C ₃ H ₄ O ₄)HSO ₄ ⁻ Cl ₂ I ⁻ (C ₅ H ₆ O ₆)HSO ₄ ⁻ (C ₂ H ₂ O ₂)(H ₂ SO ₄)HSO ₄ ⁻ (H ₂ SO ₄) ₂ HSO ₄ ⁻	C ₅ H ₇ O ₅ ⁻ IO ₃ ⁻ (HIO ₃)NO ₃ ⁻ (HIO ₃)HSO ₄ ⁻	C ₂ HO ₄ ⁻ C ₃ H ₃ O ₄ ⁻ C ₄ H ₃ O ₄ ⁻ C ₅ H ₇ O ₃ ⁻ *(C ₁₀ H ₁₄ O ₉)NO ₃ ⁻ *(C ₁₀ H ₁₆ O ₁₀)NO ₃ ⁻ *(C ₁₀ H ₁₄ O ₁₁)NO ₃ ⁻	NO ₃ ⁻ (HNO ₃)NO ₃ ⁻ C ₃ H ₄ O ₄ NO ₃ ⁻ (C ₄ H ₅ O ₃)NO ₃ ⁻ (C ₄ H ₄ O ₄)NO ₃ ⁻ (C ₅ H ₆ O ₆)NO ₃ ⁻ C ₆ H ₈ O ₆ NO ₃ ⁻ (C ₇ H ₈ O ₅)NO ₃ ⁻ (C ₈ H ₁₀ O ₅)NO ₃ ⁻ (C ₇ H ₈ O ₆)NO ₃ ⁻ (C ₈ H ₁₂ O ₅)NO ₃ ⁻ (C ₁₀ H ₁₆ O ₇)NO ₃ ⁻ *(C ₁₀ H ₁₄ O ₉)NO ₃ ⁻ *(C ₁₀ H ₁₆ O ₁₀)NO ₃ ⁻ *(C ₁₀ H ₁₄ O ₁₁)NO ₃ ⁻ (C ₉ H ₁₃ O ₁₄)NO ₃ ⁻



5. Conclusions

In this study, we investigated summertime NPF and air ion composition at a rural background site in Cyprus. NPF events were observed on approximately one-third of the measurement days (~33%), indicating that particle formation occurs regularly even under high-temperature and relatively high condensation sink conditions characteristic of the Eastern Mediterranean. The observed NPF frequency is comparable to summertime frequencies reported at Mediterranean background sites, including Finokalia in Crete (18–35 %), Helmos (27–30 %), and Sifnos (33–36 %), as well as to summertime values of approximately 20–40 % reported for boreal environments such as Hyytiälä, Finland (Aktypis et al., 2024; Dal Maso et al., 2005; Nieminen et al., 2018), despite substantial differences in atmospheric conditions. The contribution of ion-induced nucleation to total particle formation was approximately 4 %. Similar contributions have been reported in several continental environments (Manninen et al., 2010), indicating that naturally charged clusters represent only a minor fraction of the total nucleating population. Nevertheless, ambient ion measurements provide unique insight into the molecular composition of atmospheric clusters during the earliest stages of particle formation (Kirkby et al., 2016; Jokinen et al., 2018; Yan et al., 2018).

The molecular composition of ambient ions revealed a distinct diurnal evolution during NPF events. Sulfuric acid monomers, dimers and trimers, together with several mixed sulfuric acid–organic compound clusters, dominated the daytime ion population when NPF consistently occurred, whereas nighttime chemistry was characterized by HOMs clustered with nitrate. Binned PMF resolved a systematic transition from sulfuric acid dominated daytime chemistry through an iodate containing afternoon transition to HOM dominated nighttime oxidation chemistry. Despite the abundance of sulfuric acid clusters, no persistent ammonia- or amine-containing sulfuric acid clusters were detected. Only 20% of the NPF days had minute quantities of observable $\text{NH}_3(\text{H}_2\text{SO}_4)_3\text{HSO}_4^-$ contributions. This suggests either low concentrations of stabilizing base vapours or reduced stability of sulfuric acid base clusters under warm summertime conditions. Instead, several mixed sulfuric acid clusters containing small oxygenated organic compounds (e.g. $\text{C}_2\text{H}_2\text{O}_2$ and $\text{C}_3\text{H}_3\text{O}_4$) were observed during the daytime NPF period. Their direct detection under ambient atmospheric conditions provides rare molecular-level evidence that organics are associated with sulfuric acid already during the earliest stages of cluster formation. Although previous APi-TOF studies have primarily reported sulfuric acid clusters and larger HOM-containing ions in boreal environments (Bianchi et al., 2017; Schobesberger et al., 2013) our observations demonstrate that the characteristic diurnal transition from sulfuric acid to HOM-dominated ion chemistry also occurs under the substantially warmer conditions of the Eastern Mediterranean.

This study provides molecular level characterization of atmospheric ion composition during summertime NPF in the Eastern Mediterranean. These observations provide new constraints on atmospheric cluster chemistry under real atmospheric conditions and demonstrate the value of combining ambient ion (APi-TOF) and chemical ionization (CI-APi-TOF) mass spectrometry in future studies to simultaneously characterize charged molecular clusters and their neutral precursor vapours.

Data availability: The data utilized in this study are available at <https://doi.org/10.5281/zenodo.21416236> (Deot et al., 2026). The meteorological and trace gases data for AMX, from DLI, <http://www.airquality.dli.mlsi.gov.cy/>

Competing interests: The authors have no competing interests to disclose.



Author contribution: TJ conceived the study. ND and TJ performed the measurements. ND VPK, HS, KL, and TJ analysed the particle and ion data. HS and DRW supported the APi-TOF data analysis. ND and HS conducted the PMF analysis. ND, VPK and TJ prepared the manuscript with contributions from all co-authors.

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