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Observations of nanoparticle shrinkage phenomena

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Abstract. Atmospheric new particle formation (NPF) is a major source of aerosol particles in the Earth's atmosphere. However, process-level understanding of the early stages of formation and growth remains poorly represented in climate models, limiting accurate estimates of aerosol effective radiative forcing. Here, we use comprehensive observations from the Spring Particles in Cyprus (SPICY) field campaign conducted at the rural background site in Cyprus. We report new observations of atypical nanoparticle shrinkage (NPS), characterised by rapid shrinkage of sub-20 nm particles occurring in the absence of preceding NPF that typically accompanies decreasing mode diameter events. The particle size distributions exhibit a mirror image of the conventional "banana-shaped" NPF pattern, forming a distinctive "reverse-NPF" signature. We identified three NPS events during the campaign and show that this phenomenon is not driven by limited precursor gas availability, the oxidation extent of precursor vapour, or vapour scavenging by pre-existing particles. Instead, entrainment of cleaner, relatively drier air and meteorology-driven dilution, together with volatility-resolved analysis, indicate that these events are governed by atmospheric dilution and dominated by organic compounds of low and moderate volatility. Our results demonstrate that NPS events provide a previously unrecognised sink for nanoparticles, which are controlled by air-mass dynamics and organic vapour volatility.

1. Introduction

Atmospheric aerosols remain one of the largest uncertainties in quantifying effective radiative forcing and, consequently, predictions of climate change. Aerosols affect climate directly by scattering and absorbing solar radiation and indirectly by serving as cloud condensation nuclei (CCN), thereby influencing cloud properties and precipitation patterns (Twomey, 1977; Charlson et al., 1992; Rosenfeld et al., 2014; Ipcc, 2023). Atmospheric new particle formation (NPF) from low-volatility condensable vapours is prevalent throughout the troposphere (Kulmala et al., 2000; Zhang et al., 2004; Clarke, 1993), and represents the dominant global source of aerosol

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number concentrations. NPF occurs under a broad spectrum of atmospheric environments, from urban and forested sites to marine and free-tropospheric conditions (Kerminen et al., 2018; Kulmala et al., 2004; Zhao et al., 2024). Atmospheric observations indicate that 10-60% of NPF events contribute to CCN formation, enhancing their concentrations by factors that range from 0.5 to 11 at regional scales (Kerminen et al., 2018; Laakso et al., 2013; Asmi et al., 2011; Sebastian et al., 2022; Sihto et al., 2011; Rose et al., 2017; Williamson et al., 2019). Global model simulations also suggest that NPF accounts for roughly half of the tropospheric CCN population (Gordon et al., 2017; Merikanto et al., 2009; Yu and Luo, 2009). NPF events and nanoparticle growth have also been linked to severe haze episodes that exacerbate air pollution (Kulmala et al., 2022; Mishra et al., 2023; Guo et al., 2014). However, limited understanding of which condensable vapours dominate the phenomena, the initial stages of particle formation and growth, and the survival probability of nanoparticles hinder accurate representation of particle evolution from a few nanometers to CCN-active sizes (>50-100 nm) in models.

Decreasing mode diameter (DMD; also referred to as particle shrinkage) events are characterised by a characteristic evolution of particle size distributions during NPF, in which the modal diameter of particles followed by their formation and growth initially increases into the Aitken mode size range, and subsequently decreases to smaller sizes. This results in an inverted U-shaped temporal evolution of the modal diameter over time (Kamra et al., 2022; Hakala et al., 2019; Alonso-Blanco et al., 2017; Salma et al., 2016; Skrabalova et al., 2015; Cusack et al., 2013; Young et al., 2013; Yao et al., 2010; Backman et al., 2012; Hussein et al., 2020; Zhang et al., 2016; Yue et al., 2016; Kivekäs et al., 2016). As a result, DMD events can substantially suppress CCN production by preventing particles from reaching activation sizes. Cai et al. (2021) showed that limited or slow particle growth rate, together with surface tension effects, are the dominant factors constraining CCN activation during NPF events, a process that remains underrepresented in current atmospheric process-based models.

Observations across diverse regions reveal that DMD events arise from distinct mechanisms influenced by local to regional and photochemical conditions. Observations in East Asia (Hong Kong and Taiwan) suggest that DMD events are often driven by the evaporation of semi-volatile vapours under varying photochemical conditions (Yao et al., 2010; Young et al., 2013), while studies in European urban areas (Budapest, Prague, and Madrid) and densely forested environments in western Mediterranean (Montseny) attribute these events to a combination of late-afternoon reductions in condensable vapours, diminished photochemistry, evaporation of semi-volatile vapours, and increased atmospheric dilution (Salma et al., 2016; Alonso-Blanco et al., 2017; Skrabalova et al., 2015; Cusack et al., 2013). In cleaner environments, such as in the city of Toronto, limited availability of low-volatility vapours was found to inhibit particle growth, favoring DMD events compared to polluted locations like Qingdao (Zhu et al., 2014). El-Sayed et al. (2018) suggest that isoprene oxidation products under low oxides of nitrogen (NO_x) conditions can reversibly partition into condensed water in the particle phase, forming transient secondary organic aerosol (SOA) prone to evaporation. At an urban background site in Amman, Jordan, DMD events were observed on nearly 50% of NPF days and were linked to variations in particle formation and growth rates during atmospheric transport(Hussein et al., 2020). At a rural arid site in Western Saudi Arabia (Hada Al Sham), DMD events were even more frequent, occurring on 76% of NPF days(Hakala et al., 2019). Such events are driven by changes in air-mass origin (Kivekäs et al., 2016), with less-grown particles arriving at the site after larger ones, while evaporation played an insignificant role (Hakala et al., 2023). These observations indicate that

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DMD events result from a complex coupling of photochemical activity, condensing vapour availability, meteorological conditions, and air-mass history.

Here, we elucidate a new class of DMD events, which we refer to as atypical nanoparticle shrinkage (NPS) events, characterised by a rapid decrease in particle mode diameter within the sub-20 nm size range that occurs in the absence of preceding NPF typically associated with DMD events. As a result, the particle size distributions display a mirror image of the conventional "banana-shaped" NPF pattern, forming a distinctive "reverse-NPF" signature. To our knowledge, this behavior has not previously been examined in the literature. We investigate these events using comprehensive measurements carried out during the Spring Particles In CYprus (SPICY) field campaign at the rural background site, Cyprus Atmospheric Observatory - Agia Marina Xyliatou (CAO-AMX) in spring 2024.

2. Methods

2.1 Measurement Site

Cyprus is the third most populous island in the Mediterranean. The SPICY field campaign was conducted from 1 April to 15 June 2024 at the rural background site CAO-AMX (35.038692º N, 33.057850º E, 532 m a.m.s.l., Fig. 1), with a focus on processes linking direct emissions to CCN formation in an environment characterised by frequent NPF events (annual median > 58%) (Deot et al., 2025; Baalbaki et al., 2021). The campaign included an extensive suite of instruments for measuring condensing vapours, trace gases, VOCs, as well as particle size distributions and CCN concentrations. The CAO-AMX site is integrated into several international monitoring networks, including EMEP, GAW, ACTRIS, AERONET, and E-PROFILE, and is collocated with one of the nine stations of the Cyprus air quality monitoring network operated by the Department of Labour Inspection (DLI). Further details of CAO-AMX and other CAO network sites are available in Jokinen et al. (2026). The CAO-AMX site is characterised by a Mediterranean climate, with hot, dry summers and mild, wet winters. In spring, the median (range) values of temperature, relative humidity, solar radiation, and rainfall are 17.8 ºC (2.4 - 37.6 ºC), 51.2 % (7.6 - 99.2 %), 448.4 W m^{-2} (10.5 - 1027.4 W m^{-2}), and 3.1 mm (1.2 - 24.6 mm), respectively (Jokinen et al., 2026). Anthropogenic emissions around the CAO-AMX site are minimal, with the nearest major cities located ~35 km northeast (Nicosia) and ~50 km southeast (Larnaca) of the site.

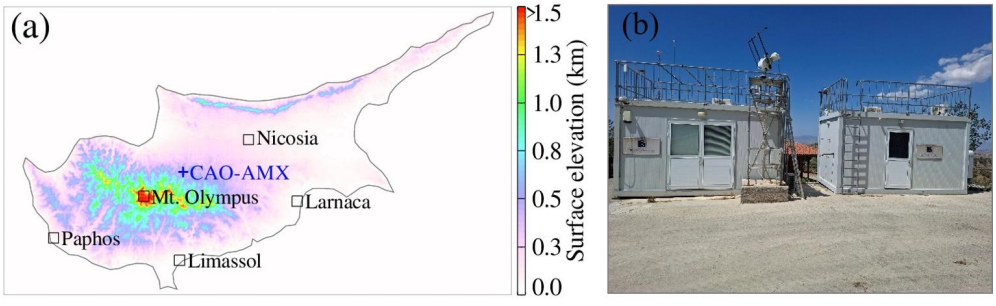

Figure 1: (a) Surface elevation map of Cyprus, showing the location of the CAO-AMX measurement station (blue marker) in the foothills of the Troodos Mountains. Major cities (Paphos, Limassol, Larnaca, and Nicosia) and Mount Olympus (the highest point in Cyprus) are indicated by open square boxes. Elevation data are taken from the U.S. Geological Survey global digital elevation model (DEM) with a horizontal resolution of 30 arc seconds

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(approximately 1 km; GTOPO30). (b) Image of the CAO-AMX station, consisting of two containers housing advanced instrumentation for condensing vapours, trace gases, and aerosols.

2.2 Instrumentation and ancillary data

2.2.1. Multi-scheme chemical IONisation Atmospheric Pressure interface Time-Of-Flight mass spectrometer (MION-APi-TOF)

The APi-TOF (Tofwerk AG, Switzerland) is a high-resolution mass spectrometer capable of identifying ambient atmospheric ions and naturally occurring ion clusters with high mass accuracy and resolving power. The APi-TOF was equipped with a second-generation MION-2 inlet (Karsa Ltd., Finland), which allows rapid switching of reagent ions based on the chemical ionisation (CI)-inlet principle and ambient naturally charged ions (Rissanen et al., 2019) . In this campaign, the MION-2 inlet was operated exclusively in negative polarity and operated with a 15-minute temporal resolution, switching between ambient ion mode for anion measurements and nitrate-based CI-mode for neutral compounds. The MION-APi-TOF sampled ambient air at 0.8 liter per minute (lpm) through a critical orifice (0.3 mm diameter). The sampled air was ionised in the MION source, and subsequently the gas-phase ions were transferred to the TOF chamber, where the charged compounds were separated according to their mass-to-charge ratio (m/z) with a resolving power of about 9000 Th/Th. Data analysis was performed using the MATLAB-based *tofTools* package developed by Junninen et al. (2010). The C_{sat} values for individual HOMs were calculated based on their molecular composition using the parametrisation by Mohr et al. (2019).

2.2.2 Proton Transfer Reaction Time-of-Flight mass spectrometer (PTR-ToF-MS)

Online measurements of VOCs were conducted using a high-resolution PTR-ToF-MS 4000 (Ionicon Analytik GmbH, Austria) following the methodology described by Desservettaz et al. (2023). The drift tube was operated at E/N = 128 Td, 2.2 mbar, 630 V, and 85 °C. Ambient air was sampled at 0.21 lpm through a heated 2-m long PEEK line with a PTFE filter, minimizing losses by using inert materials (PTFE, PEEK, glass). Mass spectra was recorded every 30 s and averaged to 10 minutes. Background signals were acquired every two hours with VOC-free air from a platinum catalyst, and calibrations were performed bi-weekly with certified standards (Apel-Riemer Environmental, USA). Data were processed with the Ionicon Data Analyzer (IDA) and matched against the GLOVOCS database (Yáñez-Serrano et al., 2016), yielding ~76 reliably quantified VOCs after applying detection limits, calibration corrections, and quality filtering. Formaldehyde was excluded due to humidity-related detection artefacts(Vlasenko et al., 2010).

2.2.3 *Nano condensation nucleus counter (nCNC)*

The A11 nCNC (Airmodus Oy, Finland) is composed of a particle size magnifier (PSM; Model A10) coupled with a CPC (Model A20) (Vanhanen et al., 2011). The sample inlet tube was approximately 60 cm long. The PSM was operated in scanning saturator flow mode (0.1 – 1.3 lpm), corresponding to a cutoff diameter of ~1.1 - 2.5 nm. To minimize artefacts, the setup included an inlet system performing background (zero) checks three times daily (12 minutes each, equivalent to three full scans) and a core sampling piece to reduce line losses of sub-3 nm particles (Baalbaki et al., 2021). A dilution with dry particle-free air behind the inlet head was employed to ensure the dew point temperatures < 20 °C, following ACTRIS standard procedures for in-situ aerosol sampling and analyses (Lehtipalo et al., 2022).

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2.2.4 Neutral Cluster and Air Ion Spectrometer (NAIS)

Ion and particle size distributions were measured using a NAIS (Airel Ltd., Estonia). The NAIS measures ion and naturally charged particle size distributions in the diameter range of 0.8–42 nm (109 size bins obtained from 25 electrometers) (Mirme and Mirme, 2013; Manninen et al., 2016), with measurements performed in both positive and negative polarities. The total particle size distribution was measured by charging aerosols using a corona charger, followed by size separation and detection carried out by two multichannel electrical mobility analyzer columns operated in parallel. 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). Further details of the NAIS setup at CAO-AMX are available in *Deot et al. (2025).*

2.2.5 Scanning Mobility Particle Sizer (SMPS)

The SMPS is composed of a TSI 3083 long differential mobility analyzer (DMA) and a TSI 3789 CPC. It measures particle size distribution between 10 and 777 nm (61 size bins). The aerosol and sheath flows were checked weekly and set to 1.0 and 4.8 lpm, respectively. The SMPS sampled ambient air using a 3-m long sample inlet tube. Aerosol sample drying was achieved using a Nafion dryer (typically RH < 40%), following the ACTRIS standard procedures for in situ aerosol sampling and analyses, and charge neutralization was achieved using a TSI radioactive (^{85}Kr) neutralizer (model 3077). The raw data from the SMPS were inverted using TSI's Aerosol Instrument Manager (AIM, version 11.5) software. The nano-DMA SMPS, consisting of a TSI 3085 DMA and a TSI water-based 3786 CPC, was also operated simultaneously, with a sample flow rate of 0.3 lpm. The nano-DMA SMPS sampled ambient air using a 2-m long sample inlet tube. It measures particle size distribution between 3.1 and 105.5 nm (99 size bins). Additionally, a TSI butanol-based standalone 3750 CPC (D50 = 7 nm) was also operated, with a sample flow rate of 1 lpm.

2.2.6 Aethalometer

BC mass concentrations were measured using a seven-wavelength aethalometer (AE33, Magee Scientific, USA) at 1-minute resolution. Ambient aerosol was sampled through a PM_1 inlet at 5 lpm after drying with a Nafion dryer. BC concentrations were derived at 880 nm using a mass absorption cross-section of 7.77 $m^2\ g^{-1}$ (Petzold et al., 2013). This wavelength was chosen for calculating BC concentration because absorption by other aerosols is negligible at this wavelength (Drinovec et al., 2015). The aethalometer uses Teflon-coated glass fiber tape, and aerosols are collected onto two parallel spots for optical absorption measurements, which provide real-time compensation for the filter loading effect.

2.2.7 Ceilometer

The Vaisala Ceilometer CL51, in operation since 2021 and part of the E-PROFILE network , provides vertical profiles of aerosols and clouds (Münkel and Roininen, 2010). It employs an eye-safe InGaAs diode-laser lidar (910 ± 10 nm, 110 ns pulses, 6.5 kHz) in a vertical or near-vertical direction, detecting aerosols and clouds from ~300 m to 15 km with 10 m resolution. Backscatter profiles are processed with the BL-VIEW software, which applies a gradient method with a "cloud and precipitation filter" to remove spurious signals (Münkel and Roininen,

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2010; Emeis et al., 2007). The software combines gradient and idealized backscatter approaches to automatically estimate planetary boundary layer (PBL) height at 16s temporal and 10 m vertical resolution. In this study, we retained Level 3 boundary layer height data with a "good" quality control index.

2.2.8 Ancillary observations

Collocated (within a 50 m radius) trace gas measurements, including NO_x, CO, and O_3, as well as meteorological parameters, including air temperature, relative humidity, solar radiation, wind speed, and wind direction, were obtained from the national air quality monitoring network operated by the DLI of Cyprus. O_3 was measured with UV photometric monitors (Ecotech 9810B). CO was measured with non-dispersive IR (NDIR) spectroscopy monitors (Ecotech 9830B). NO_x was measured with the chemiluminescence technique. Solar radiation was measured with a Kipp & Zonen CMP22 solar irradiance sensor. Further details of DLI instrumentation are available in Vrekoussis et al. (2022). In addition, aerosol optical depth (AOD) was obtained from the AERONET sun photometers operated at the CAO-AMX site. All data are reported in Coordinated Universal Time (UTC). Local time in Cyprus is UTC+3 during daylight saving time (Eastern European Summer Time, late March to late October). Only quality-assured and quality-controlled datasets, based on standard procedures and instrument-specific data quality flags, were used in this study. Air-mass backward trajectories were computed using NOAA's Air Resources Laboratory (ARL), Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) PC-version model, version 4.4 (Draxler and Rolph, 2010; Stein et al., 2015), incorporating meteorological data from the Global Data Assimilation System (GDAS) with a spatial resolution of $0.25^{o} \times 0.25^{o}$ and a temporal resolution of 1 hour (Draxler and Rolph, 2010). Two-day hourly backward air-mass trajectories, initialized at 500 m above the ground level, were used in this analysis. We further used ERA5, a fifth-generation reanalysis produced by the European Centre for Medium-Range Weather Forecasts (ECMWF), 0.25° gridded data of vertical velocity and specific humidity at hourly resolution on pressure levels from 1000 hPa to 550 hPa (Hersbach et al., 2023).

3. Results

3.1 Identification of event types

Event types were classified by visual inspection of particle size distribution contour plots (Dal Maso et al., 2005). A day was classified as an NPF event when a pronounced increase in particle number concentrations was observed in the nucleation size range (<25 nm), followed by continuous growth of these particles to larger diameters. Such events typically extend over spatial scales of hundreds of kilometers and persist for 1–2 days, and are therefore characterised as regional NPF events(Kulmala et al., 2004). NPF events are often accompanied by a decrease in particle mode diameter from the Aitken size range during the afternoon hours, a phenomenon referred to as DMD (or shrinkage event) in the literature. As described earlier, we also observed atypical events characterised by a rapid decrease in particle mode diameter within the sub-20 nm size range that occurred in the absence of the preceding particle formation and growth typically associated with DMD events, referred to as atypical NPS. Such atypical behavior has previously been reported at our site (Baalbaki et al., 2021) and in Pune, India (Kamra et al., 2022), but the underlying processes were not examined, and similar events have not been reported elsewhere. Notably, the particle size distribution contour plot during an NPS event displays a mirror image of the conventional "banana-shaped" NPF pattern, appearing as a distinctive "reverse-NPF" shape. A day without the onset of a new particle mode in the nucleation size range is classified as a non-event day. A day that could not be

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classified into any of the above categories was classified as an unclear day. A day without a valid full-day measurement, due to instrument maintenance, troubleshooting, or occasional power outages, was classified as a "nodata" day. Figure S1 shows the temporal evolution of median particle size distributions from nano Condensation Nucleus Counter (nCNC), Neutral Air Ion Spectrometer (NAIS; negative polarity ions and particles), and scanning mobility particle sizer (SMPS), while Fig. S2 shows typical examples of NPF, DMD, NPS, unclear, and non-event days.

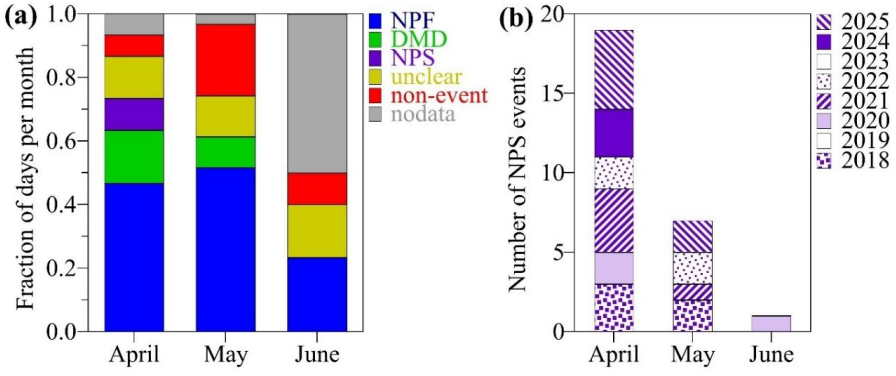

Figure 2: (a) Monthly fraction of days classified into different event types during the SPICY campaign (1 April – 15 June 2024). (b) Total number of NPS event days identified from 2018 to 2025. There are no measurements during 2019 and 2023.

Figure 2a shows the monthly fraction of days classified into different event types during the SPICY campaign. NPF events accounted for the largest fraction of days, occurring on 44 of the 76 campaign days (57.9%). Of these, 37 days were classified as conventional NPF events (48.7%), while 8 days were identified as DMD events (10.5%). Atypical NPS events were observed on 3 days (3.9%). non-events and unclear days accounted for 12 (15.8%) and 13 days (17.1%), respectively. There were no valid measurements on 3 days (3.9%) during the campaign. Here, we present a two-day consecutive case study consisting of an NPF event on 6 April followed by an NPS event on 7 April. This consecutive case was selected to minimize variability associated with regional meteorological conditions and to enable a detailed analysis of the physicochemical mechanisms underlying the NPS phenomenon. In addition, two further consecutive NPF-NPS cases (11-12 April and 20-21 April) are presented in the Supplementary Information (Figs. S3-S7), and all three cases are summarised below. Long-term measurements of particle size distributions at the CAO-AMX site indicate that NPS is a commonly observed phenomenon (Fig. 2b), highlighting the importance of the comprehensive SPICY observations for elucidating its underlying mechanisms.

3.2 Elucidating an atypical nanoparticle shrinkage event

Figure 3 shows the median diurnal variation of particle size distributions and aerosol properties during the consecutive NPF event on 6 April, followed by an NPS on 7 April. A pronounced sub-3 nm particle burst (nCNC; Fig. 3a), followed by a typical "banana-shaped" aerosol growth pattern in the particle size distribution (Fig. 3b-d), was observed during the NPF event. In contrast, the NPS event exhibited a rapid decrease in nanoparticle mode

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diameter (<20 nm) without any preceding evidence of particle formation or subsequent growth. During the NPF event, an approximately 2-hour time lag between the peak number concentrations of sub-3nm (nCNC) and >7 nm CPC particles clearly indicate particle growth, whereas the nearly simultaneous peaks observed during the NPS event suggest the absence of such growth (Fig. 3e). While the concentrations of ions with diameters 2.0 - 2.3 nm, indicative of local intermediate ion (2 - 7 nm) formation (Tuovinen et al., 2024), increased sharply during the NPF event, they remained negligible during the NPS event (Fig. 3e). This indicates the absence of local NPF processes during the NPS event. The number concentrations of nucleation mode ($N_{10\text{-}25nm}$) and total particles ($N_{10\text{-}777nm}$) were comparable on 6 and 7 April. In contrast, both the total condensation sink ($CS_{10\text{-}777nm}$) and particulate matter with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$) were approximately twofold lower during the NPS event than during the NPF event (Fig. 3f). While a lower CS generally favors the growth of small clusters or newly formed particles by reducing vapour losses to pre-existing particles (Kulmala et al., 2001), it also reflects cleaner atmospheric conditions with limited availability of condensing vapours. Consistently, black carbon (BC) mass concentrations were slightly lower during the NPS event, indicating a minimal influence from local anthropogenic sources such as fossil fuel combustion or biomass burning. This further suggests the absence of nearby or advected nanoparticle sources reaching the site. Two other consecutive NPF-NPS cases showed a similar temporal evolution of aerosol properties (Figs. S3 and S4), except for higher pre-existing particle concentrations during the NPS event in the 10-11 April case.

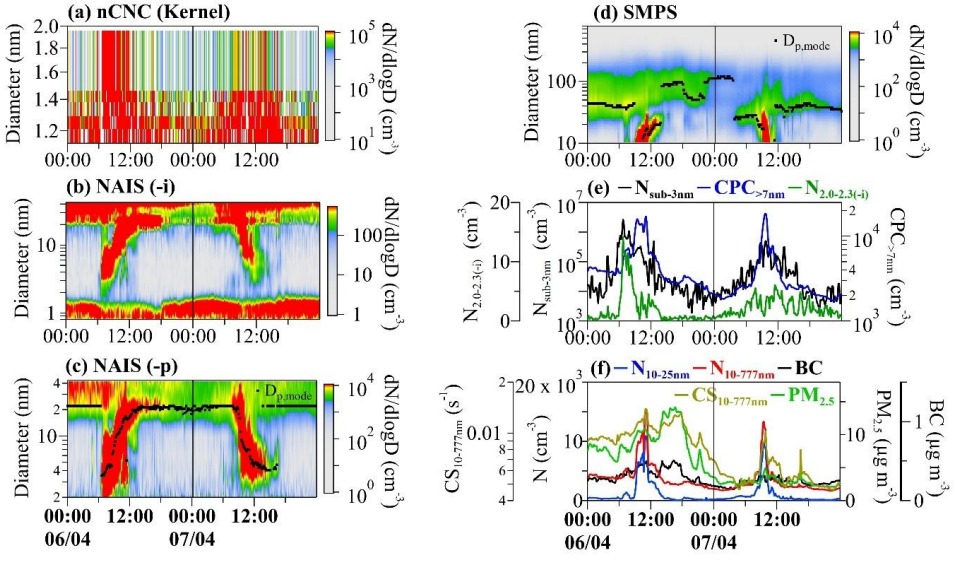

Figure 3: Median diurnal variation of aerosol properties during the consecutive NPF (6 April) and NPS (7 April) events. Particle size distributions measured by (a) nCNC, (b-c) NAIS negative polarity ions and particles, and (d) SMPS. Concentrations of (e) negative polarity ions in the 2.0-2.3 nm size range measured by NAIS ($N_{2\text{-}2.3(\text{-}i)}$), sub-3nm particles measured by nCNC ($N_{sub\text{-}3nm}$), and >7nm particles measured by CPC ($N_{>7nm}$); (f) nucleation mode ($N_{10\text{-}25nm}$) and total particle ($N_{10\text{-}777nm}$) number concentrations from SMPS, total condensation sink ($CS_{10\text{-}}$

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$_{777nm}$), and particulate matter of aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) and black carbon (BC) mass concentrations.

From a meteorological perspective (Fig. 4), the diurnal variation in temperatures was similar on 6 and 7 April, with mean temperatures of 20.1 °C and 19.9 °C (averaged over 6-12 UTC) during the NPF and NPS events, respectively. Relative humidity was also comparable, with mean values of 30.4% during the NPF event and 30.7% during the NPS event. Wind speeds were similar for both events, although the wind direction differed; it was northeasterly on 6 April and northwesterly on 7 April during the 6-12 UTC period (Fig. 4b). The boundary layer height and solar radiation (Fig. 4c) likewise show no substantial differences between the two events. Overall, local meteorological conditions were largely comparable between the NPF and NPS events, with wind direction being the notable difference. A similarly consistent temporal evolution of meteorological parameters was observed for the two additional consecutive NPF-NPS cases (Fig. S5), indicating comparable meteorological conditions across events and suggesting that the observed differences between NPF and NPS were not primarily driven by local meteorology.

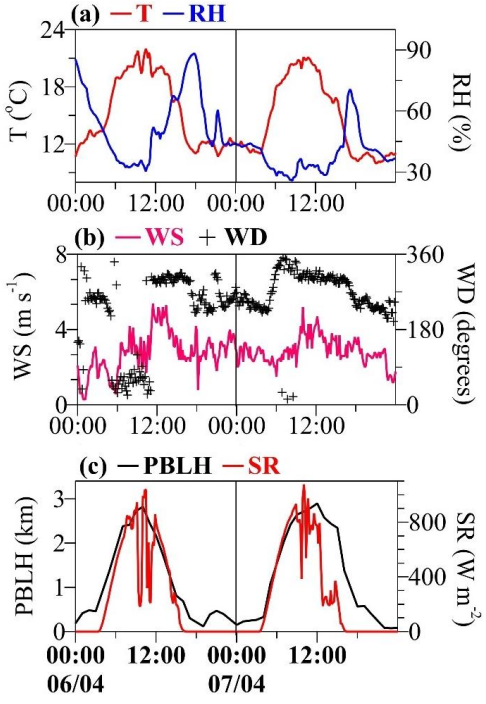

Figure 4: Median diurnal variation of meteorological parameters during the consecutive NPF-NPS events (6-7 April). (a) air temperature (T) and relative humidity (RH), (b) wind speed (WS) and wind direction (WD), and (c) planetary boundary layer height (PBLH) and solar radiation (SR).

From a chemical process perspective, volatile organic compounds (VOC) mixing ratios and their early oxidation products were examined to identify the dominant chemical regimes associated with the NPF and NPS events. VOCs at the CAO-AMX site are controlled by local temperature and continental transport(Garg et al., 2025), with

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long-term observations indicating the onset of local biogenic emissions in mid-March. Mixing ratios of benzene and toluene (anthropogenic tracers), as well as biogenic tracers such as isoprene and monoterpenes (MTs), were comparable between the two days (Fig. 5a). Methyl vinyl ketone (MVK) and methacrolein (MACR), first-generation OH-initiated oxidation products of isoprene, showed similar behavior on both days; accordingly, the (MVK+MACR)/isoprene ratio, a proxy for the extent of isoprene oxidation, was also comparable (Fig. 5b). Plant chamber studies (Kiendler-Scharr et al., 2009) and field observations (Kanawade et al., 2011) have shown that isoprene can suppress biogenic NPF at ground level, thereby dampening aerosol cooling effects (Lee et al., 2016), with the extent of suppression depending on the relative contributions of isoprene- and monoterpene-derived carbon. To compare the relative OH-initiated oxidation rates of isoprene and MTs, we calculated the ratio ($k_{Iso\text{-}OH}$ × [Isoprene]) / ($k_{MT\text{-}OH}$ × [MT]), where $k_{Iso\text{-}OH} = 1.01\times10^{-10}$ cm^3 $molecules^{-1}$ s^{-1} and $k_{\alpha\text{-}pinene\text{-}OH} = 5.36\times10^{-11}$ cm^3 $molecules^{-1}$ s^{-1} represent the reaction rate coefficients of isoprene and alpha-pinene with OH radical, respectively (Finlayson-Pitts and Pitts, 2000). This ratio was slightly higher during the NPF event (Fig. 5b), indicating an isoprene-dominated OH-reactivity regime that favors the formation of more volatile, lower O/C oxidation products, which are less favorable for biogenic NPF. In contrast, the lower ratio observed during the NPS event rules out isoprene-driven suppression of biogenic NPF under those conditions; nevertheless, NPF did not occur on 7 April. Curtius et al. (2024) showed that organonitrates formed from OH-initiated oxidation of isoprene in the presence of nitrogen oxides drive NPF in the upper troposphere over the Amazon. Consistently, concentrations of isoprene-derived hydroxy hydroperoxy nitrate ($C_5H_{10}O_4(ONO_2)$) measured by MION-APi-TOF were comparable on both days, indicating that the initial stages of isoprene oxidation proceeded similarly during the two events (Fig. 5c). In contrast, peak concentrations of isoprene-derived dinitrates ($C_5H_{10}O_2(ONO_2)_2$) were slightly higher during the NPF event than during the NPS event, indicating a stronger influence of high-NO_x driven chemistry during the NPF event. This interpretation is supported by the reactive nitrogen compounds (NO_y) concentrations, which were also slightly higher during the NPF event than during the NPS event (Fig. 5c). Further, acetaldehyde (C_2H_4O), a marker of atmospheric oxidation, also exhibited similar mixing ratios on both days, indicating comparable overall oxidation levels (Fig. 5c).

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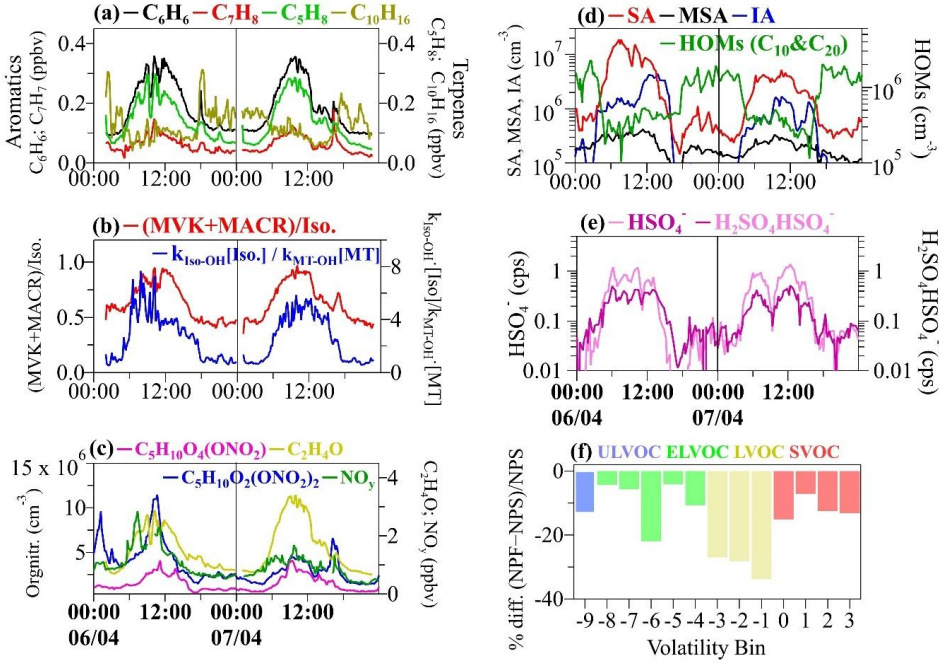

Figure 5: Median diurnal variation of aerosol precursors and condensing vapours during the consecutive NPF-NPS events observed during 6-7 April. (a) mixing ratios of benzene (C_6H_6), toluene (C_7H_8), isoprene (C_5H_8), and monoterpenes ($C_{10}H_{16}$);(b) the ratio of the sum of methyl vinyl ketone (MVK) and maethacrolein (MACR) to the isoprene [(MVK+MACR)/Iso.] and the relative OH reaction rates of isoprene and MTs ($k_{Iso\text{-}OH}$·[Iso]/$k_{MT\text{-}OH}$·[MT]); (c) organonitrates [$C_5H_{10}O_4(ONO_2)$ and $C_5H_{10}O_2(ONO_2)_2$], acetaldehyde (C_2H_4O) and reactive nitrogen compounds (NO_y); (d) concentrations of sulfuric acid (SA), methyl sulfonic acid (MSA), iodic acid (IA), and highly oxygenated organic molecules (HOMs, C_{10} & C_{20}); (e) ambient ion mode signals of bisulfate monomer (HSO_4^-) and dimer ($H_2SO_4HSO_4^-$) in counts per second (cps); (f) percentage difference in organic vapour concentrations (molecules cm^{-3}) between the NPF and NPS events as a function of volatility bin, expressed as $\log_{10}$ of the effective saturation concentration, $\log_{10}$(C*). Negative percentage differences indicate higher organic vapour concentrations during the NPS event. ULVOC, ELVOC, LVOC, and SVOC denote ultra-low volatile organic carbon, extremely low volatile organic carbon, low volatile organic carbon, and semi-volatile organic carbon, respectively.

We next examined neutral aerosol precursors, including sulfuric acid (SA), methanesulfonic acid (MSA), iodic acid (IA), and HOMs, measured using the nitrate mode of the MION-APi-TOF. SA concentrations were approximately fivefold higher during the NPF event than during the NPS event. Nevertheless, SA concentrations during the NPS event (~4 × 10^6 molecules cm^{-3}) were sufficient to trigger NPF under atmospherically relevant ammonia mixing ratios of 100 parts per trillion by volume (pptv) and dimethylamine mixing ratios of 40 pptv (Kirkby et al., 2011; Kürten et al., 2018). Concurrently, HOMs concentrations increased during the NPF event, but decreased during the NPS event (Fig. 5d), whereas MSA and IA concentrations were comparable on both days (Fig. 5d). Negative ion measurements from APi-TOF further showed that concentrations of bisulfate monomers

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(HSO_4^-) and dimers ($H_2SO_4HSO_4^-$) were similar on both days (Fig. 5e), indicating comparable levels of ionised SA clusters in the ambient air. No indication of higher multimers or ammonia-containing clusters was detected.

Chamber experiments have demonstrated that initial nanoparticle growth is dominated by extremely low-volatility organics (ELVOC; approximately $C^*<10^{-4.5}$ µg m^{-3}, volatility bin <-4), whereas growth at larger sizes is increasingly supported by more abundant, slightly higher-volatility vapours in the low-volatility organic compounds (LVOC) range ($C^* \approx 10^{-4.5}$ - $10^{-0.5}$; -4 > volatility bin < 0) as the Kelvin barrier falls with increasing diameter (Tröstl et al., 2016). We, therefore, examined differences in organic vapour concentrations relevant to early cluster formation and subsequent growth between the two event types (Fig. 5f). Organic vapour concentrations were higher during the NPS event than during the NPF event across all volatility bins (Fig. 5f), indicating that particle growth was not limited by the availability of condensable vapours. Instead, these conditions point to an environment less favorable for stable initial cluster formation and subsequent growth, and more favorable to net evaporation of condensed material. Dilution of the air-mass lowers organic aerosol concentrations and thus the absorptive capacity of the particle phase, shifting gas-particle equilibrium toward the vapour phase and promoting the evaporation of particle-phase semi-volatile organic carbons (SVOCs) (Pankow, 1994). This repartitioning can cause aerosol mass to decrease more rapidly than expected from dilution alone, reflecting the dynamic response of SVOCs to changing ambient conditions (Donahue et al., 2014). Under ambient conditions, SVOCs partition between the gas and particle phases, whereas LVOCs and ELVOCs exist primarily, or entirely, in the particle phase (Barsanti et al., 2017). Consistent with this framework, the observed reduction in HOM concentrations (Fig. 5d, Figs. S6-S7) and the absence of particle growth suggest that dilution associated with the entrainment of cleaner, drier free tropospheric air into the boundary layer played a key role during NPS events. The similar chemical environment (Fig. 5; Figs. S6-S7) and local meteorological conditions (Fig. 4; Fig. S5), together associated with the contrasting particle evolution (Fig. 3; Figs S3-S4), indicates that the observed differences in particle behavior were not driven by the availability of condensing vapours, their oxidation products, local atmospheric conditions, or vapour scavenging rates.

We therefore hypothesize that atmospheric dilution creates conditions that are less favorable for particle formation and subsequent growth in the atmosphere. Air-mass trajectory analysis shows distinct transport pathways and air-mass characteristics between the two cases, particularly on 6 and 7 April. During the NPF event, air masses travelled slowly near the ocean surface and largely remained within the PBL, whereas during the NPS event, they descended from free tropospheric levels to near-surface at the passage of the Western Taurus mountain range (Fig. S8). Similar contrast in air-mass history between NPF and NPS events was observed for the 10-11 April case (Fig. S9), while the air-mass history was comparable and mainly oceanic between the NPF and NPS events for the 20-21 April case (Fig. S10). Hakala et al. (2023) proposed that overlapping nucleation modes from distinct nearby source regions may be advected to a site. Here, however, we observed both homogenous and contrasting air-mass source regions, indicating that long-range advection of nucleation mode particles is unlikely to explain the observed NPS events. Vertical velocity and specific humidity were further examined to characterise the dynamic state of the lower atmosphere (Fig. S11). Stronger low-level subsidence, accompanied by lower specific humidity (i.e., drier air), was evident in the upwind region of the measurement site on NPS event days. Such enhanced vertical mixing and entrainment of cleaner, drier free tropospheric air are indicative of atmospheric dilution and

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are consistent with the observed lower near-surface aerosol mass (Fig. 6a-b) and reduced columnar loading (Fig. 6c), as well as lower NO_x (Fig. 6d) and carbon monoxide (CO; Fig. 6e) concentrations and higher ozone (O_3) mixing ratio (Fig. 6f).

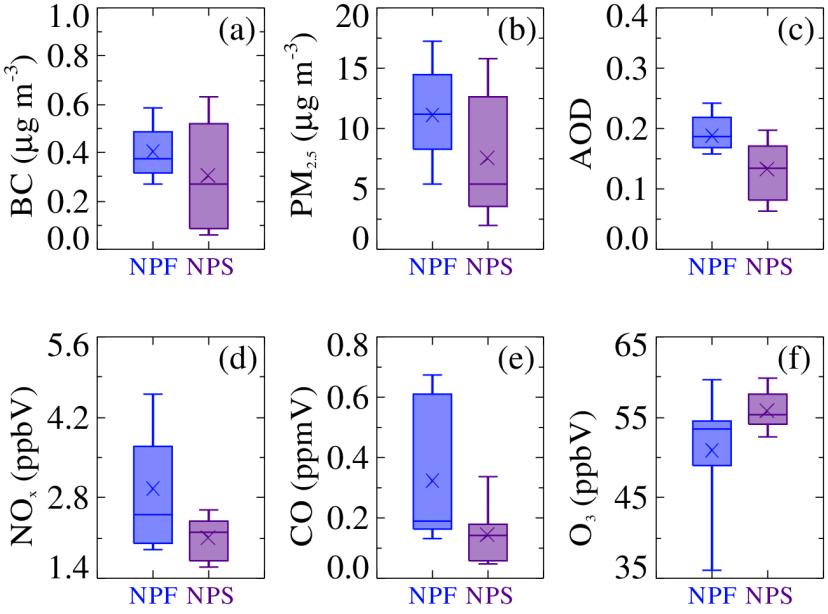

Figure 6: Key differences in aerosol and gases during observed NPF (6, 10, and 20 April) and NPS (7, 11, and 21 April) event days. Box–whisker plots show (a) black carbon (BC) mass concentrations, (b) particulate matter with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$), (c) aerosol optical depth (AOD), (d) oxides of nitrogen (NO_x), (e) carbon monoxide (CO), and (f) ozone (O_3). Data correspond to the 06–12 UTC period. The cross symbol indicates the mean, the horizontal line indicates the median, the bottom and top of the box indicate the 25th and 75th percentiles, and the bottom and top of the whisker indicate the 10th and 90th percentiles.

The origin of the observed nanoparticles during NPS event days can be constrained into two physically consistent scenarios: the particles were either transported from nearby anthropogenic sources or formed locally but failed to grow. The low BC mass concentrations (Fig. 6a), together with reduced NO_x (Fig. 6d) and CO (Fig. 6e) levels, indicate that contributions from local primary emissions are unlikely. Although the number concentrations of 2.0 - 2.3 nm ions, indicative of local intermediate ion formation, were only slightly lower during the NPS event than during NPF event (Fig. S12a), concentrations of sub-3nm neutral particles (Fig. S12b) and the number concentrations of 2.5 - 7 nm and 7 - 25 nm particles were significantly lower during NPS events than during NPF events (Fig. S12c-d). The reduced concentrations of 2.0 - 2.3 nm ions during NPS events can be explained by reduced levels of condensing vapours (Fig. 5d) and a lower condensation sink (Fig. S12e), and lower pre-existing particles (Fig. 6b). The higher concentrations of LVOCs and SVOCs observed during the NPS event (Fig. 5f) would, in principle, be expected to condense onto existing particles and promote growth; however, condensation onto the smallest particles is likely suppressed by the strong curvature (Kelvin) effect. This inhibition limits nanoparticle growth despite the availability of condensable material, preventing particles from growing to larger

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sizes. Consistent with this interpretation, the low total particle volume concentrations observed during NPS event days (Fig. S12f), together with elevated LVOC and SVOC concentrations (Fig. 5f), point to evaporation of condensed material primarily driven by atmospheric dilution.

4. Conclusions

This study presents new evidence of nanoparticle shrinkage observed at the rural background site (CAO-AMX) in Cyprus. These events are characterised by a rapid reduction in particle diameters within the sub-20 nm size range, with the particle size distributions contour plot displaying a mirror image of the conventional "banana-shaped" NPF pattern. To our knowledge, such behavior has not previously been elucidated; therefore, we hereby expand the current phenomenology of atmospheric aerosol evolution.

Using a suite of instruments that enabled comprehensive gas- and particle-phase measurements, we conclude that NPS events are not driven by limited gas-phase nucleating precursors, oxidation intensity, or enhanced scavenging of condensing vapour by pre-existing particles. SA concentrations during NPS events were sufficient to trigger NPF under atmospherically relevant boundary layer conditions, while key VOC mixing ratios and oxidation rates were comparable between NPF and NPS cases. Moreover, the limited local cluster formation, low black carbon and NO_x concentrations, and a distinct air-mass source region rule out local primary emissions as the origin of the observed sub-20 nm particles. Having ruled out the above-mentioned mechanisms, we conclude that NPS events were driven by atmospheric dilution associated with the entrainment of cleaner, drier air from the free troposphere, leading to reduced concentrations of highly oxygenated organic molecules and a relative enhancement of organic compounds with low to moderate volatility, together with higher O_3 mixing ratios. Volatility-resolved analysis further indicates that, although ULVOCs and ELVOCs concentrations were slightly higher during NPS events, particle formation and subsequent growth were inhibited. Under these conditions, reversible partitioning of low- and semi-volatile organic compounds favors net evaporation, leading to rapid particle shrinkage.

Our findings highlight a previously unrecognised sink for nanoparticles occurring under conditions favorable for particle formation and growth. This can negatively impact the CCN budget, thereby damping the aerosol-induced radiative cooling effect.

Code and data availability

Time-synchronised 10-minute median 1D and 3D data variables are available at Zenodo (https://doi.org/10.5281/zenodo.18685092). Ceilometer data can be viewed at https://e-profile.eu/. ERA5 reanalysis data are publicly available from Copernicus Climate Change Service (C3S) Climate Data Store (CDS) at https://cds.climate.copernicus.eu/datasets (last access: January 2025). AERONET aerosol optical depth data are available publicly to download from https://aeronet.gsfc.nasa.gov/ (last access: October 2025). Surface elevation data from the U.S. Geological Survey Earth Resources Observation and Science (EROS) Center is available publicly to download from https://www.usgs.gov/centers/eros/science/usgs-eros-archive-digital-elevation-global-30-arc-second-elevation-gtopo30 (last access: 22 October 2024)

The IGOR Pro (V9) data analysis tool is available from the corresponding author upon reasonable request.

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Supplement link

The link to the supplement will be included by Copernicus, if applicable.

Author contributions

VPK, MP, FM, GB, EB, JS, and TJ designed the experiments. ND, AC, MvdB, AP, MP, SB, and CS carried them out. VPK, MC, AC, CL, AP, RB, MP, and AG analyzed the data. VPK prepared the manuscript with contributions from all co-authors.

Competing interests

The author declares that they have no conflict of interest.

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