



Online single-particle characterization of ice-nucleating particles (INPs) at the High Altitude Research Station Jungfraujoch (3580 m a.s.l.), Switzerland

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Abstract. Ice-nucleating particles (INPs) modulate the phase, radiative properties, and lifetime of clouds, yet direct observational evidence linking single aerosol particles to atmospheric ice nucleation (IN) remains challenging because of the low abundance of INPs. We sampled ambient aerosols by coupling an INP counter to a single-particle mass spectrometry (SPMS) via a pumped counter-flow virtual impactor at Jungfraujoch during wintertime. Ice crystal residuals (ICRs) sampled under conditions representative of mixed-phase and cirrus cloud formation and total aerosol sampled directly by the SPMS were chemically characterized. Mineral dust (MD) dominated the ICR population under both cloud regimes despite its minor contribution to ambient aerosol particles by number. Two distinct MD subclasses associated with local construction activities (MD-K/Al/Fe) and transported sources (MD-Ca) were identified. MD-K/Al/Fe contributed substantially to the INP population in the cirrus cloud regime, demonstrating that local anthropogenic activity can influence atmospheric INPs populations. In contrast, sulfate, organics, nitrate, and biomass burning aerosol (BBA) particles contributed only a minor fraction of ICRs despite dominating the ambient aerosol population, indicating low IN activity under the investigated conditions. However, Pb signatures were enriched in BBA ICRs, suggesting enhanced IN activity of Pb-containing BBA particles, whereas no corresponding enrichment was observed for Pb-containing MD-K/Al/Fe particles. Sporadic meteoric particles detected during stratospheric-intrusion events indicate a potential contribution of stratospheric aerosols to tropospheric primary IN. Limited in-cloud observations suggest that natural cloud processing may modify the IN activity of MD-K/Al/Fe and BBA particles.



1 Introduction

Previous studies have identified several classes of aerosol particles with prescribed composition and size capable of acting as ice-nucleating particles (INPs) through different ice nucleation (IN) modes across various cloud regimes, including primary particles from natural sources, such as mineral dust (MD), sea spray aerosol (SSA), biological aerosol, biomass burning aerosol (BBA), and volcanic ash. Anthropogenic sources include black carbon (BC), brown carbon, fly ash, and secondary organic aerosols (Pruppacher and Klett, 1997; Kanji et al., 2017; Burrows et al., 2022). Among all INP types, MD particles originating from surface soil and deserts are considered the most abundant natural INPs due to their high atmospheric loading (Kok et al., 2023, and references therein) and their ability to facilitate IN at temperatures lower than -12 to -15 °C via both immersion freezing and deposition IN (e.g., Archuleta et al., 2005; Möhler et al., 2006; Atkinson et al., 2013; Welti et al., 2014; Ullrich et al., 2017, 2019). Biological aerosol particles exhibit IN activity between -2 and -12 °C in immersion freezing mode (e.g., Möhler et al., 2007; Hoose et al., 2010; Conen et al., 2015; Seifried et al., 2021; Gratzl et al., 2025), although their low abundance at cloud-forming altitude might limit their overall importance for ice formation in clouds. The organic component of SSA particles has also been reported as effective INPs in both immersion freezing (e.g., DeMott et al., 2016; Wolf et al., 2020a) and deposition IN modes (e.g., Wolf et al., 2021; Patnaude et al., 2024). BBA particles contribute significantly to global aerosol loading (Schill et al., 2020) but show variable IN activities (e.g., Grawe et al., 2016; Jahn et al., 2020; Jahl et al., 2021; Chen et al., 2025). Anthropogenic aerosol particles such as BC particles generally exhibit poor IN activity in immersion freezing but could initiate IN effectively below -38 °C (e.g., Mahrt et al., 2018; Nichman et al., 2019; Mahrt et al., 2020a, b; Zhang et al., 2020; Kanji et al., 2020; Gao et al., 2022a; Gao and Kanji, 2022; Gao et al., 2022b), though the freezing mechanism might be pore condensation freezing rather than deposition IN (e.g., Marcolli, 2014, 2017; Mahrt et al., 2018; Marcolli, 2020; Zhang et al., 2020; Marcolli et al., 2021). Secondary aerosol particles have been identified as effective INP via deposition IN in the cirrus cloud (CC) regime largely depending on their phase state (e.g., Abbatt et al., 2006; Murray et al., 2010; Zhang et al., 2018a; Kilchhofer et al., 2021; Wang et al., 2022). Nevertheless, most laboratory investigations use proxy particles that may not fully represent ambient aerosols after atmospheric aging.

Online field measurements of INPs at cloud-forming altitudes can capture transient meteorological and aerosol conditions that may not be represented by laboratory surrogates. Several previous studies at the High Altitude Research Station Jungfraujoch (JFJ; 46.330° N, 7.590° E, 3580 m a.s.l.) have attempted to infer INP species and sources by correlating online atmospheric INP number concentration (N_{INP}) with ambient aerosol particle physicochemical properties, such as particle number and/or mass concentration within specific size ranges, along with aerosol optical properties and chemical composition obtained from parallel measurements (e.g., Chou et al., 2011; Boose et al., 2016; Lacher et al., 2017, 2018, 2021; Brunner et al., 2022) or reanalysis data (e.g., Brunner et al., 2021). These ground-based observations corroborate laboratory findings that MD particles dominate ambient INP populations at JFJ, especially during Saharan dust events (SDEs; Collaud Coen et al., 2004, 2026). In addition to mineral dust, long-range transported SSA has also been proposed as a potentially important INP source at JFJ.



Despite the ≈ 350 km distance between JFJ and the nearest Mediterranean coastline, Boose et al. (2016) and Lacher et al. (2017, 2021) suggested that long-range transported SSA may also act as effective INPs based on the cloud water sampling containing marine organics. It is also suggested that the biological material associated with MD (e.g., Prenni et al., 2009; Conen et al., 2011; O'Sullivan et al., 2014; O'Sullivan et al., 2015; Steinke et al., 2016; O'Sullivan et al., 2018) and SSA (e.g., Burrows et al., 2013; Wilson et al., 2015; Wilbourn et al., 2020) contributes to the relatively superior IN activity of these aerosol classes.

To resolve these uncertainties, single-particle characterization techniques are essential for identifying which individual ambient aerosol particles initiate atmospheric IN. A commonly adopted approach involves characterizing the physicochemical properties of cloud ice particle residuals (IPRs) after separating ice particles from cloud droplets and interstitial aerosol particles using counter-flow virtual impactor inlets (Ogren et al., 1985; Boulter et al., 2006; Kulkarni et al., 2011; Hiranuma et al., 2016). This method has been applied both onboard research aircraft (e.g., Cziczo et al., 2004; Froyd et al., 2010; Cziczo et al., 2013) and at ground-based research stations (Mertes et al., 2007; Richardson et al., 2007; Cozic et al., 2007, 2008a; Cziczo et al., 2009; Kamphus et al., 2010; Ebert et al., 2011; Worringen et al., 2015; Kupiszewski et al., 2016; Schmidt et al., 2017; Eriksen Hammer et al., 2018; Cornwell et al., 2019; Karlsson et al., 2021; Lacher et al., 2021; Adachi et al., 2022b). Aircraft-based measurements typically focus on CC IPRs to avoid interference from cloud droplets in mixed-phase cloud (MPC), while ground-based studies employ specialized inlets (Mertes et al., 2007; Kupiszewski et al., 2015) to investigate MPC IPRs. Common techniques for IPR characterization include online single-particle mass spectrometry (SPMS; e.g., Cziczo et al., 2006; Murphy, 2007; Cziczo et al., 2017; Jacquot et al., 2024) and offline electron microscopy (e.g., Ebert et al., 2002). However, offline electron microscopy analysis requires long sampling times up to days due to the scarcity of ambient INPs and IPRs, which can lead to mixing of different air masses in one sample and hinder IPR composition assignment to specific clouds and sources (Ebert et al., 2011; Adachi et al., 2022b).

Extensive single-particle characterization of IPRs in MPCs using both SPMS and electron microscopy has been carried out at JFJ during multiple CLOUD–Aerosol Characterization Experiment (CLACE) and Ice Nuclei research UNIT (INUIT) campaigns between 2004 and 2017 (Mertes et al., 2007; Cozic et al., 2007, 2008a; Cziczo et al., 2009; Kamphus et al., 2010; Ebert et al., 2011; Worringen et al., 2015; Kupiszewski et al., 2016; Schmidt et al., 2017; Eriksen Hammer et al., 2018; Lacher et al., 2021). IPRs are consistently enriched in MD and depleted in sulfate, organics, and nitrate (SON) particles compared to ambient aerosol particle number concentration (Mertes et al., 2007; Kamphus et al., 2010; Ebert et al., 2011; Worringen et al., 2015; Kupiszewski et al., 2016; Schmidt et al., 2017; Eriksen Hammer et al., 2018; Lacher et al., 2021), indicating a preference for MD particles to be IN scavenged and therefore a high IN activity via immersion freezing. Metallic particles, particularly those containing Pb, are also frequently detected in IPRs (Cziczo et al., 2009; Kamphus et al., 2010; Ebert et al., 2011; Worringen et al., 2015; Schmidt et al., 2017). Mixing state analysis suggests that internal mixing with Pb, regardless of its chemical form, may enhance the immersion freezing activity of ambient aerosol particles (Cziczo et al., 2009; Ebert et al., 2011; Welti et al., 2019). The role of BC in MPC IPR remains debated. While enrichment of BC mass has been observed using filter-based light transmission (Cozic et al., 2007, 2008a; Mertes et al., 2007) and electron microscopy (Ebert et al., 2011; Eriksen Hammer et al., 2018), Kupiszewski et al. (2016) reported depletion of BC by number in IPRs. The apparent



contradiction is attributed to interference from MD particles in aerosol optical absorption measurements, whereby enhanced MD absorption led to an overestimation of BC mass in IPRs. Although IPRs embedded in cloud ice particles might originate from the INPs that initiated IN, their presence can also result from coagulation with residuals formed during ice growth through rimming, scavenging, and cloud chemistry processes (Ebert et al., 2011; Lloyd et al., 2015; Eriksen Hammer et al., 2018). Despite extensive IPR measurements, direct observational evidence identifying the aerosol particles responsible for initiating ice formation in natural clouds remains sparse.

Both IPRs and ice crystal residuals (ICRs) are aerosol particles remaining after ice sublimation. However, IPRs collected from natural clouds may reflect subsequent cloud processing, ICRs are generated under controlled freezing conditions and are therefore directly linked to the INPs that initiated ice formation. A bottom-up method enables direct characterization of ambient INP by first exposing INPs to predefined thermodynamic conditions to produce pristine ice crystals, followed by sublimation and single-particle characterization of the resulting ICRs using electron microscopy (Chen et al., 1998; Worringer et al., 2015; Conen et al., 2026) or SPMS (Cziczo et al., 2003, 2009; DeMott et al., 2003; Richardson et al., 2007; Gallavardin et al., 2008; Corbin et al., 2012; Baustian et al., 2012). Online SPMS measurements of deposition ICRs have been conducted at the high altitude Storm Peak Laboratory (3220 m a.s.l., 40.455° N, 106.744° W; Cziczo et al., 2003; DeMott et al., 2003; Richardson et al., 2007; Baustian et al., 2012) and at an urban site in Toronto (Corbin et al., 2012). More recently, Cornwell et al. (2019) conducted online SPMS analysis of immersion mode INPs at a coastal site in California. These online SPMS studies consistently reveal that MD and metallic particles are the major component of ambient INP, regardless of the freezing modes, sampling location, and measurement technique, in agreement with the offline ICR findings from aircraft (Chen et al., 1998) and ground-based (Worringer et al., 2015) platforms. Beyond these common particle classes, ICR composition reflects contributions from diverse aerosol sources. Urban combustion aerosols, including BC and BBA, have been identified as effective deposition INPs (Corbin et al., 2012, $T = -34 \pm 1$ °C and $RH_w \approx 98$ %), whereas marine aerosols, particularly organic nitrogen-enriched SSA, have been associated with efficient immersion freezing (Cornwell et al., 2019, $T = -31$ °C and $RH_w = 105$ %). Moreover, the importance of Pb-containing particles remains uncertain (Cziczo et al., 2009). Reported Pb fractions in ICRs range from ≈ 20 % at Storm Peak Laboratory (Baustian et al., 2012, $T = -43 \pm 1$ °C and $RH_w = 92$ %) to only 1.5 % at JFJ (Worringer et al., 2015, $T = -22$ °C and RH_w between 92 and 145 %), while several studies detected little or no Pb enrichment in ICR (Cziczo et al., 2003; DeMott et al., 2003; Richardson et al., 2007; Corbin et al., 2012). Collectively, these studies highlight that direct chemical characterization and source attribution of individual ambient INPs remain limited across different freezing modes and atmospheric environments.

To address these uncertainties, we deployed an online IN-SPMS setup at JFJ to characterize the chemical composition of ICRs, identify ambient INP sources, and evaluate the IN activity of individual particle classes under thermodynamic conditions representative of MPC and CC formation.



115 2 Methods

2.1 Site description

The measurement campaign was conducted from January 22 to February 14 2024 at the Sphinx Observatory (46.330° N, 7.590° E, 3580 m a.s.l.) at the JFJ. The experimental setup is shown in Fig. 1. Ambient air was sampled through a total aerosol inlet with a 40 μm cut-off diameter at wind speeds up to 20 m s^{-1} (Weingartner et al., 1999). The inlet surfaces were heated to
120 35 °C to prevent ice accumulation and to evaporate freshly formed hydrometeors. Consequently, the sampled air comprised interstitial aerosol particles, cloud droplet residuals, and IPRs. This total aerosol inlet is part of the continuous aerosol monitoring program within the Global Atmospheric Watch and the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS; Mohr and Gysel-Beer, 2024; Laj et al., 2024).

The research station also provides one-hour averaged meteorological and trace gas data through the SwissMetNet meteorological network operated by MeteoSwiss and the Swiss National Air Pollution Monitoring Network operated by Empa.
125 Detailed descriptions of meteorological and trace gas monitoring at JFJ can be found in Appenzeller et al. (2008) and Hueglin et al. (2024), respectively. In addition, ^{222}Rn has been measured at JFJ since 2009 by the University of Basel and serves as an indicator of boundary layer intrusions (BLIs; Griffiths et al., 2014). The probability of JFJ being in free troposphere (FT) and BLI occurrence was calculated following the method proposed by Brunner et al. (2021), which combines the ^{222}Rn (Griffiths
130 et al., 2014) and the number concentration of particles with a mobility diameter $d_m \geq 90 \text{ nm}$ ($N_{\geq 90}$; Herrmann et al., 2015; Lacher et al., 2018, 2021) data.

2.2 Online single-particle characterization of INP

Single-particle characterization of INPs was performed by coupling the Automated Horizontal Ice Nucleation Chamber (HINC-Auto; Brunner and Kanji, 2021) to the SPMS Particle Analysis by Laser Mass Spectrometry-Next Generation (PALMS-NG, Jacquot et al., 2024) and Ultra High Sensitivity Aerosol Spectrometer (UHSAS; DMT) via a Pumped Counterflow Virtual
135 Impactor (PCVI; BMI Model 8100; Boulter et al., 2006), shown as the dashed line in Fig. 1. Ambient N_{INP} and aerosol particle chemical composition measurement was performed before and after each single-particle characterization of INPs, as indicated by the solid line in Fig. 1.

INPs were first activated into pristine ice crystals within HINC-Auto under prescribed thermodynamic conditions. Down-
140 stream of the chamber, the ice crystals were separated from liquid droplets and interstitial aerosol particles by the PCVI, owing to the higher inertia of ice crystals. After sublimating the ice crystals at room temperature, the resulting ICRs were introduced into PALMS-NG and UHSAS for chemical composition and size analysis.

2.2.1 INP activation in HINC-Auto

HINC-Auto is a continuous flow diffusion chamber instrument made of two flat parallel copper-coated aluminum plates whose
145 inner walls are coated with ice. The temperature of each plate is controlled independently to create a supersaturation along

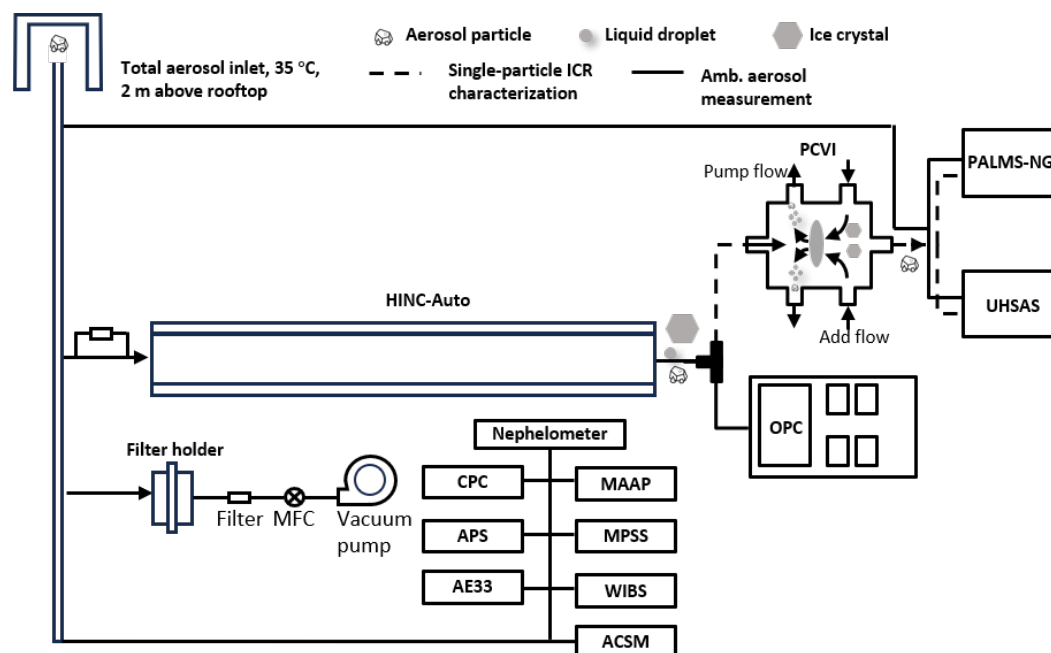


Figure 1. Schematic of the sampling and experimental setup. The dashed line indicates the single-particle ICR characterization experiment, and the solid line indicates the ambient aerosol characterization experiment when Particle Analysis by Laser Mass Spectrometry–Next Generation (PALMS-NG) and Automated Horizontal Ice Nucleation Chamber (HINC-Auto) measure in parallel. Acronyms: pumped counter-flow virtual impactor (PCVI); Ultra-High Sensitivity Aerosol Spectrometer (UHSAS); Optical Particle Counter (OPC); Mass Flow Controller (MFC); Aethalometer (AE33); Multi Angle Absorption Photometer (MAAP), Condensation Particle Counter (CPC); Aerodynamic Particle Sizer (APS); Mobility Particle Size Spectrometer (MPSS); Wideband Integrated Bioaerosol Sensor (WIBS); Aerosol Chemical Speciation Monitor (ACSM).

the chamber centerline lamina. To minimize the impact of convection, the top plate of HINC-Auto is warmer than the bottom plate. Due to its horizontal design, HINC-Auto has a built-in aerosol cutoff size of $\approx 2.5 \mu\text{m}$. More details of HINC-Auto can be found in Brunner and Kanji (2021).

To simulate both MPC and CC conditions in this study, HINC-Auto was operated at $T = -30 \text{ }^\circ\text{C}$ and $RH_w = 104 \%$ from January 22 to February 2, and at $T = -40 \text{ }^\circ\text{C}$ and $RH_w = 90 \%$ from February 3 to February 14, as indicated by the arrows in Fig. 2(a). The sample flow rate of HINC-Auto was $0.28 \text{ std L min}^{-1}$ (where std L denotes standard liters referenced to $0 \text{ }^\circ\text{C}$ and 1013.25 hPa), surrounded by $2.55 \text{ std L min}^{-1}$ particle-free sheath air. During the campaign, the sampled air passed through a particle filter for 5 minutes after every 15 minutes of measurements to quantify the instrument signal to noise.

2.2.2 Ice crystal separation

The PCVI uses a particle-free flow in the direction opposite to the motion of the ice crystals to separate them from liquid droplets and interstitial aerosol particles (Boulter et al., 2006). Ice crystals do not follow the streamlines due to their larger

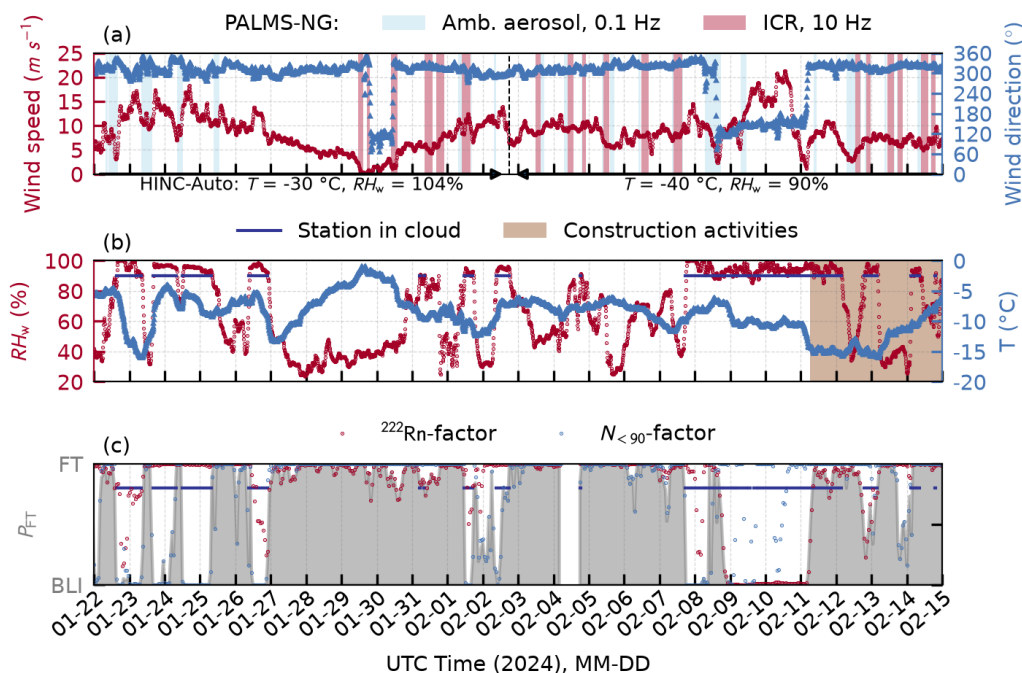


Figure 2. Times series of (a) wind speed and direction, (b) RH_w and T , and (c) the probability of FT and BLI occurrence at JFJ. In panel (a), blue and red shaded periods indicate PALMS-NG measurement periods dedicated to ambient single-particle aerosol measurements and ICRs characterization at different sampling frequencies, respectively. In panel (b), blue lines indicate periods when the station was in cloud, while brown shading denotes measurement periods affected by construction activities. In panel (c), grey shading indicates periods when the site was within the FT.

inertia and can penetrate the virtual impaction plane, shown as the gray oval in Fig. 1, for further analysis. The liquid droplets and aerosol particles follow the streamlines and get pumped out from the upstream part of PCVI, as schematically showed by the arrows in Fig. 1.

160 In the single-particle characterization of INP setup shown in Fig. 1, the INP input flow rate of PCVI was $2.83 \text{ std L min}^{-1}$, matching the HINC-Auto output flow (Brunner and Kanji, 2021). The PCVI output flow was $0.47 \text{ std L min}^{-1}$, determined by the PALMS-NG ($0.40 \text{ std L min}^{-1}$) and UHSAS ($0.07 \text{ std L min}^{-1}$). The add flow and pump flow of the PCVI were set to $1.20 \text{ std L min}^{-1}$ and $3.56 \text{ std L min}^{-1}$, respectively, to yield a cutoff size around $4 \mu\text{m}$, consistent with the size threshold for ice crystal detection in HINC-Auto. A similar flow rate setup was used by Corbin et al. (2012). To minimize the impact of artifact

165 particles that might penetrate the virtual impaction plane of the PCVI (Pekour and Cziczo, 2011; Cziczo et al., 2017), only ICRs with vacuum aerodynamic diameter (d_{va}) $\geq 0.6 \mu\text{m}$ are considered as representative INPs in this study. This corresponds to a geometric diameter of $\geq 0.32 \mu\text{m}$, assuming a shape factor of 1.4 and a particle density of 2.6 g cm^{-3} (Hiranuma et al., 2015; Kanji et al., 2019). Particles with $d_{va} < 0.6 \mu\text{m}$ may include ICRs in addition to the breakthrough particles that bypassed



the PCVI separation. Consequently, the applied size threshold provides a conservative estimate of ambient INP abundance at
170 JFJ.

2.2.3 Online single-particle ICR characterization

PALMS-NG is a bipolar SPMS that can measure the d_{va} and both the positive and negative mass spectra of a single aerosol particle in the size range from 0.1 to 4 μm in real-time (Jacquot et al., 2024). d_{va} measurement is realized by detecting light scattering incidence and measuring the time-of-flight when an aerosol particle consecutively crosses the path of two
175 405 nm laser beams. The mass spectra are obtained using a single-step laser desorption and ionization technique, i.e., the aerosol particle is ablated and ionized by a 193 nm pulsed excimer laser. Ions are analyzed with a bipolar time-of-flight mass spectrometer, revealing the chemical composition of the detected particle. PALMS-NG allows for a rapid classification of the particles to pre-defined classes. For a full description of PALMS-NG, please refer to Jacquot et al. (2024) and Shen et al. (2025).

180 In this study, mass spectra obtained from PALMS-NG are classified into MD, SON, BBA, and Other particle classes including meteoric (MT) and SSA, based on the characteristic peaks using the algorithm reported in Froyd et al. (2019) and Shen et al. (2025). The (EF) is defined as the ratio of the number fraction of a given particle class in ICRs to that in ambient aerosol particles within the same size range ($0.6 \mu\text{m} \leq d_{va} \leq 4 \mu\text{m}$ Cziczo et al., 2003; Ebert et al., 2011; Schmidt et al., 2017). $EF > 1$ indicates enrichment. During the campaign, a few detected ICRs only had size information but did not produce distinguishable
185 spectra and were also classified as Other particles, which was probably related to the excimer laser gas degrade (Murphy, 2007; Kamphus et al., 2010). Excimer laser gas was refilled on January 30 and February 11 to optimize ablation and ionization efficiency and maximize the meaningful signal. The sample flow rate of PALMS-NG was 0.40 std L min^{-1} . A high efficiency particulate air filter was placed at the PALMS-NG inlet before and after each measurement period to ensure clean instrument background during the campaign. PALMS-NG measurement periods are highlighted as red and blue shadings in Fig. 2a.

190 The UHSAS was connected in parallel to PALMS-NG at the PCVI exit to characterize the ICR size distribution. UHSAS is an optical-scattering, laser-based aerosol particle spectrometer system for sizing particles in the 0.055 to 1.0 μm range. The sample flow rate of the UHSAS was 0.07 std L min^{-1} .

2.3 Online ambient aerosol particle characterization

When operated in ambient aerosol characterization mode (solid lines in Fig.1), all instruments were connected to the total
195 aerosol inlet in parallel. Comprehensive descriptions of aerosol monitoring at JFJ are available elsewhere (Baltensperger et al., 1997; Fröhlich et al., 2015; Herrmann et al., 2015; Bukowiecki et al., 2016; Collaud Coen et al., 2026; Masoom et al., 2026; Mazzini et al., 2026; Weng et al., 2026). A brief overview of the instruments deployed is provided here. Sub-micron particle number size distribution was measured by a Mobility Particle Size Spectrometer (MPSS, Wiedensohler et al., 2018). The MPSS operated at 0.65 std L min^{-1} with a sheath-to-sample flow ratio of 5, covering ambient aerosol particles with mobility
200 diameter (d_m) from 0.011 to 0.790 μm . An Aerodynamic Particle Sizer (APS; model 3321; TSI Inc.) measured the number size distribution for ambient aerosol particles with aerodynamic diameter (d_a) from 0.542 μm to 19.81 μm using a sample



flow rate of $0.65 \text{ std L min}^{-1}$. Total ambient aerosol particle number concentration (N_{CPC} , $d_m > 0.01 \mu\text{m}$) was monitored by a Condensation Particle Counter (CPC; model 3772; TSI Inc.) with a sample flow rate of $0.65 \text{ std L min}^{-1}$. A Wideband Integrated Bioaerosol Sensor (WIBS; Droplet Measurement Technologies) quantified the number concentration of fluorescent particles with d_a between 0.5 and $30 \mu\text{m}$ (N_{WIBS}) at a sample flow rate of $0.20 \text{ std L min}^{-1}$. d_a was converted to d_m by assuming the effective density of wintertime ambient aerosol particles at JFJ to be 1.2 g cm^{-3} , and the shape factor to be unity (Cozic et al., 2008b; Chou et al., 2011). A detailed discussion on different diameter definitions can be found in DeCarlo et al. (2004).

Real-time non-refractory PM1 ($d_a < 1.0 \mu\text{m}$) mass concentration and chemical composition (m_{PM1}) were measured by an Aerosol Chemical Speciation Monitor (ACSM) equipped with a Time of Flight Mass Spectrometer (TOF-ACSM; Aerodyne Inc.) operating at $0.07 \text{ std L min}^{-1}$. Equivalent black carbon mass concentration (m_{BC}) was monitored separately by an Aethalometer (AE33, Magee Sci. Inc.) and a Multi Angle Absorption Photometer (MAAP, model 5012; Thermo, Inc.). Single-scattering albedo and absorption Ångström exponent (SSAAE) was calculated following Collaud Coen et al. (2004, 2026) using light scattering measurements from a nephelometer (model 3565; TSI Inc.) and light absorption measurements from the AE33, and was used as an indicator of SDEs.

2.4 Lagrangian source analysis

Two three-dimensional Lagrangian kinematic models, namely the Lagrangian particle dispersion model FLEXible PARTicle dispersion model (FLEXPART; Stohl et al., 2005; Henne et al., 2010, 2016) and Lagrangian parcel model LAGRangian Analysis TOol (LAGRANTO; Wernli and Davies, 1997; Miltenberger et al., 2013; Sprenger and Wernli, 2015) were used to investigate air-mass history and aerosol transport pathways. The Consortium for Small-scale Modeling (COSMO) version of both FLEXPART and LAGRANTO uses the high-resolution COSMO output with horizontal resolution of $\approx 7 \text{ km}$, hourly time resolution, and 79 vertical levels, as meteorological input to resolve the complex orographic Alpine wind field. Once the air parcel leaves the COSMO domain, FLEXPART and LAGRANTO use European Centre for Medium-Range Weather Forecasts (ECMWF) Integrating Forecasting System (IFS) and Reanalysis v5 (ERA5) as meteorological input, respectively. The ERA5 input used for LAGRANTO has a horizontal resolution of $0.5^\circ \times 0.5^\circ$, 98 vertical levels, and hourly temporal resolution (Hersbach et al., 2020).

LAGRANTO was run every 6 hours to compute 7-day back trajectories from 10-km equidistant points within a 15 km radius around JFJ, using 6 pressure levels evenly distributed between 600 and 700 hPa for each point. Potential vorticity (PV), and RH_w were tracked along each trajectory to identify stratospheric intrusion (Cui et al., 2009). Aerosol properties, including 3-D species-specific optical depth, were traced using collocated Copernicus Atmosphere Monitoring Service (CAMS) reanalysis fields (Inness et al., 2019).

FLEXPART was run at 3-hour intervals and released particles at 3470 m a.s.l. to account for the topographic smoothing around JFJ in the model. Source-receptor relationship was computed to determine total and boundary-layer source sensitivities.

3 Results and discussion

3.1 Overview

During the campaign, air masses arriving at JFJ originated predominantly from the northwest (300° – 350°) with wind speeds up to 22.3 m s^{-1} (Fig. 2a). The surrounding Alpine topography and glaciers channel local airflow at JFJ predominantly along a northwest-southeast axis. Consequently, the measured wind direction reflects local flow conditions rather than the large-scale transport pathways of the air masses (e.g., Brunner et al., 2021). Depending on the prevailing meteorological conditions, these air masses may have acquired SSA and MD particles while traversing the Atlantic Ocean, the Mediterranean Sea, or the Sahara before reaching JFJ (Figs. A1–A5). There were two periods characterized by predominantly southeasterly wind, i.e. from 20:50 UTC on January 29 to 10:00 UTC on January 30, and from 15:20 UTC on February 8 to 03:50 UTC on February 11 (Fig. 2a). During these periods, the air masses passed over the Po Valley and were likely influenced by anthropogenic emissions (Fig. A5). Ambient temperature ranged between -0.7 and -16.2°C , with a campaign average of -8.8°C . Owing to its altitude, JFJ was in FT air masses over 60 % of the campaign, consistent with previous winter observations (Herrmann et al., 2015), while sporadic midday BLIs were also observed (Fig. 2c). Periods of elevated RH_w , frequently approaching 100 %, generally coincided with BLIs, indicating in-cloud conditions ($RH_w \geq 90$ % in this study) and potential precipitation.

During ambient cloud events (Fig. 3b), reduced N_{CPC} , which is dominated by sub-micron particles (Fig. 3b and e), suggests effective in-cloud scavenging. Correspondingly, non-refractory PM1 mass concentration and m_{BC} (Fig. 3a) follow N_{CPC} closely, with $m_{\text{PM1}} < 0.1 \mu\text{g m}^{-3}$ during cloudy periods. Nevertheless, large amount of in-cloud PM1 appeared on January 24 and February 2 (dominated by sulfate) and on February 12 (dominated by nitrate and organics). These PM1 particles might be linked to domestic heating and vehicle emission transported upslope from the valley. The particle volume size distribution (Fig. 3f), dominated by super-micron particles, aligns well with $N_{>500}$, $N_{>1000}$, $N_{\text{WIBS-Total}}$, and $N_{\text{WIBS-Fluo}}$ (Fig. 3c and d). On-site construction activities associated with the construction of a new tunnel between February 11 and February 14 (Fig. 2b) likely contributed additional super-micron particles through rock drilling, excavation, material handling, and machinery emissions, while other occasional super-micron aerosol particles, for example on January 29, were likely associated with SDEs (Fig. A1), consistent with negative SSAE shadings in Fig. 3e and f.

Single-particle INP characterization yielded 74.5 hours of data and 2248 ICR spectra during MPC and CC processing experiments at a sampling frequency up to 10 Hz (Fig. 4c, d, and g). Parallel ambient aerosol particle measurement produced 35410 spectra of ambient aerosol particles with $0.1 \mu\text{m} < d_{\text{va}} \leq 4 \mu\text{m}$ over 65 hours at a fixed sampling frequency of 0.1 Hz, including 27153 spectra collected during the non-construction period and 8257 spectra during the construction period (Fig. 4a and e). Two distinct MD subclasses were identified: particles with K/Al/Fe (MD-K/Al/Fe) and Ca (MD-Ca) signatures, respectively, with representative spectra shown in Fig. 5a.

PALMS-NG collected 25 ICRs with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ during the non-construction period (January 15 to February 11), including 15 ICRs sampled during MPC processing experiments over 27.5 hours (Fig. 4c), and 10 during CC processing experiments over 25 hours (Fig. 4d). During the construction period (February 11 to February 15), PALMS-NG collected 2223

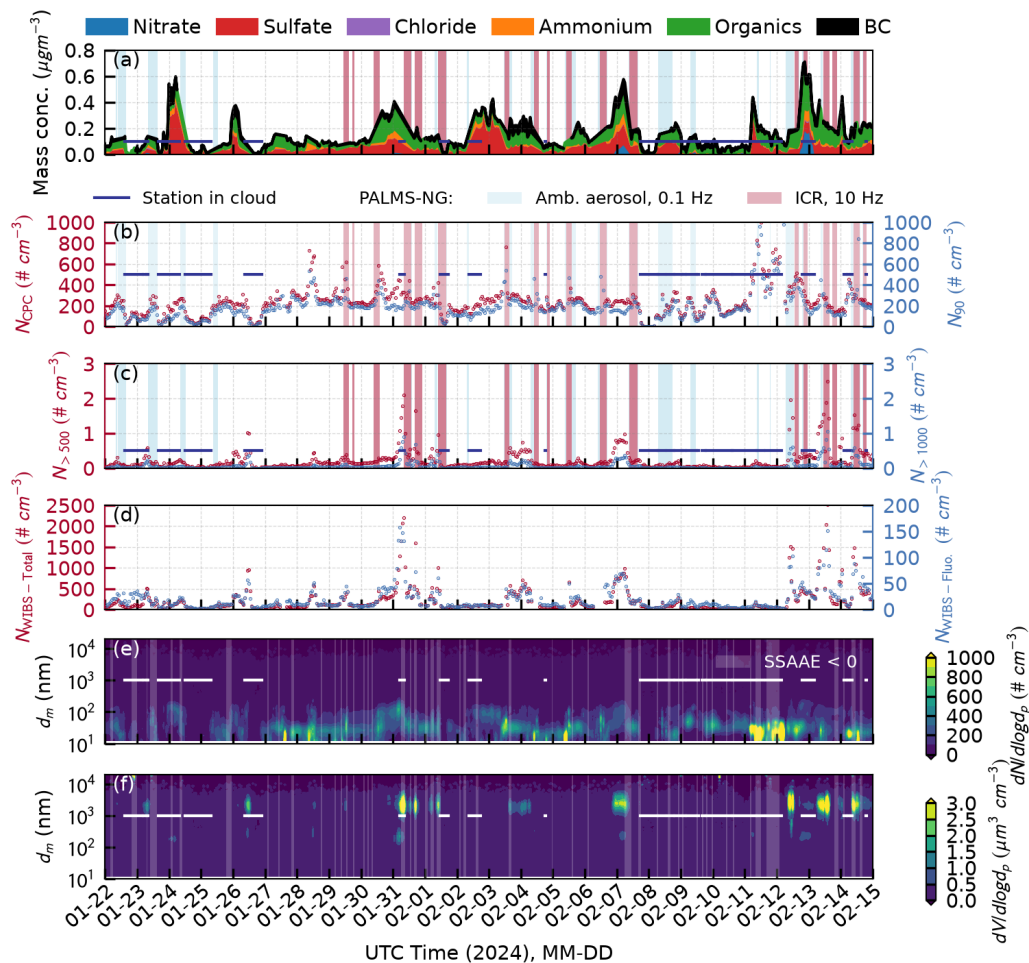


Figure 3. Time series of (a) non-refractory PM1 and BC mass concentration, (b) number concentration of total ambient aerosol particles (N_{CPC}) and particles with $d_m < 90$ nm ($N_{\geq 90}$), (c) number concentration of aerosol particles with $d_m > 500$ nm ($N_{>500}$) and 1000 nm ($N_{>1000}$), (d) total ($N_{\text{WIBS-Total}}$) and fluorescent ($N_{\text{WIBS-Fluo}}$) particle number concentration measured by WIBS, (e) particle number size distribution, and (f) particle volume size distribution. The shadings in panel (e) and (f) denote periods when SSAAE is negative, indicative of MD presence (Collaud Coen et al., 2004, 2026).

ICRs after CC processing over 22 hours, of which 78.5 % were classified as MD-K/Al/Fe particles and 4.3 % as MD-Ca particles (Fig. 4g).



3.2 IN activity of ambient INPs derived from ICR classes

270 3.2.1 SON, BBA, and SSA particles are not effective INPs

As shown in Fig. 4a and e, ambient aerosol particles sampled by PALMS-NG ($0.1 \mu\text{m} < d_{\text{va}} \leq 4 \mu\text{m}$) are dominated by SON and BBA during both the non-construction and construction periods. For larger particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$, SON (31.9 %) and BBA (32.0 %) each contribute approximately one-third of the ambient aerosol spectra during the non-construction period, together accounting for 63.9 % of all particles (Fig. 4b). During the construction period, however, the contribution of
 275 SON decreases to 14.2 % (Fig. 4f), mainly because construction activities introduce larger MD particles, which are discussed in Sect. 3.2.2.

The contributions of SON and BBA decrease substantially after cloud processing within HINC-Auto, indicating that these particle classes are strongly depleted downstream of the PCVI. During the non-construction period, the proportion of both SON and BBA decrease from ≈ 32 % in ambient aerosol particles to 6.7 % after MPC and 10.0 % after CC processing (Fig. 4b–d).
 280 A similar reduction is observed during the construction period, with the SON percentage decreasing from 14.2 % in ambient aerosol particles to 4.6 % after CC processing and BBA from 33.9 % to 8.0 % (Fig. 4f and g).

These reductions correspond to *EF*s of 0.21 after MPC processing and 0.31 after CC processing for both SON and BBA particles during the non-construction period (Table 1). A similar depletion is observed during the construction period after CC processing, with BBA exhibiting slightly stronger depletion (*EF* = 0.23) than SON (*EF* = 0.32). This pronounced depletion
 285 indicates that SON and BBA particles are inefficient INP under both immersion-freezing ($T = -30$ °C, $RH_w = 104$ %) and deposition IN ($T = -40$ °C, $RH_w = 90$ %) conditions.

BBA particles at JFJ likely originate from regional residential wood-burning for domestic heating in the Alpine region or long-range transport of wildfire plumes (Masoom et al., 2026), characterized by K^+ ($m/z = 39$) and organic fragment (e.g., $m/z = 28 \text{CO}^+$, $m/z = 44 \text{CO}_2^+$, $m/z = 24 \text{C}_2^-$, $m/z = 25 \text{C}_2\text{H}^-$, $m/z = 26 \text{C}_2\text{H}_2^-$ or CN^-) signals. The composition
 290 and light absorption measurements from the long-term observations suggest that long-range transported BBA particles mainly consist of organics internally mixed with minor fractions of inorganic salts, BC and likely also tar (amorphous carbon). Ambient SON and BBA particles commonly contain hygroscopic species such as SON and ammonium. Under MPC and CC conditions in HINC-Auto, these particles may undergo hygroscopic growth and activate into liquid droplets. These activated liquid droplets and unactivated aerosol particles are below the cutoff size of the PCVI and follow the exhaust flow and therefore are not sampled
 295 by PALMS-NG. Kamphus et al. (2010) observed even stronger depletion for SON and BBA particles in IPRs with *EF* down to 0.03 relative to ambient aerosol particles using two SPMSs. This observation agrees with laboratory results showing that freshly emitted and aged BBA particles from fuels like wood and leaves are generally ineffective as immersion-freezing INPs (e.g., Petters et al., 2009; Chou et al., 2013; McCluskey et al., 2014; Jahn et al., 2020, 2021; Chen et al., 2025).

Despite the strong depletion of SON and BBA in ICRs, it remains unclear whether inadvertant transmission within PCVI,
 300 IN or scavenging, or internal mixing with other INPs is responsible for their presence in the observed ICRs. Laboratory studies have shown that solid sulfate and nitrate particles (e.g., Abbatt et al., 2006; Baustian et al., 2010; Wang et al., 2022) as well as amorphous glassy organic particles (e.g., Berkemeier et al., 2014; Jensen et al., 2018; Zhang et al., 2018a, b; Kasparoglu

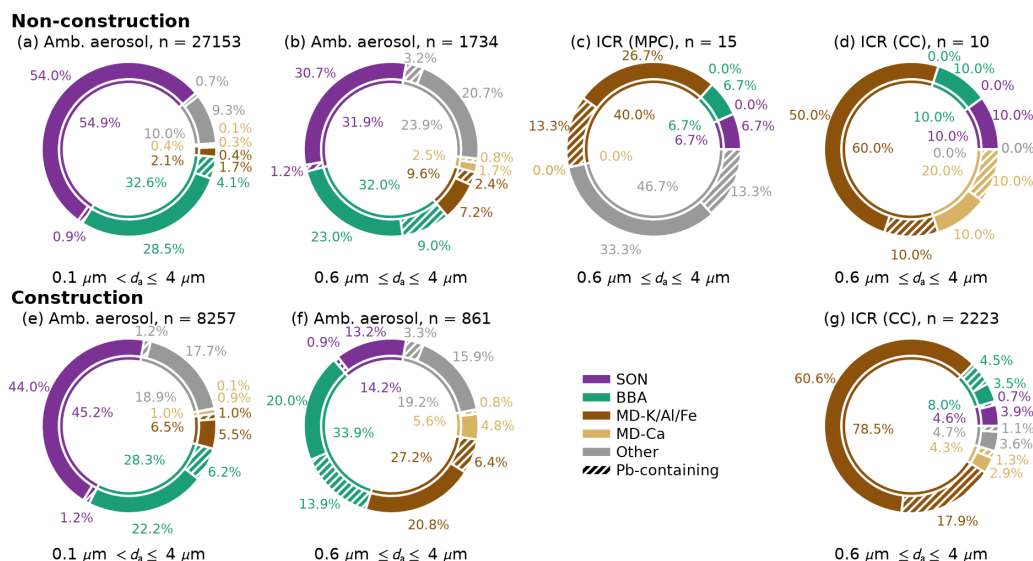


Figure 4. Number percentage of particle classes measured by PALMS-NG during (a – d) non-construction and (e – g) construction periods: (a, e) ambient aerosol particle with $0.1 \mu\text{m} < d_{va} \leq 4 \mu\text{m}$ and (b, f) $0.6 \mu\text{m} \leq d_{va} \leq 4 \mu\text{m}$, as well as ICRs with $0.6 \mu\text{m} \leq d_{va} \leq 4 \mu\text{m}$ after (c) MPC processing ($T = -30^\circ\text{C}$, $RH_w = 104\%$) and (d, g) CC processing ($T = -40^\circ\text{C}$, $RH_w = 90\%$). Hatched areas in the outer circles denote particles exhibiting Pb signatures, and numerical labels indicate number percentage values. The inner ring shows the total fraction of each particle class, whereas the outer ring separates each class into Pb-containing (hatched) and non-Pb-containing fractions; thus, the inner-ring percentage equals the sum of the corresponding outer-ring percentages for each particle class. SON – sulfate, organics, and nitrate, BBA – biomass burning aerosol, MD – mineral dust.

Table 1. *EFs* of PALMS-NG particle classes in ICRs relative to ambient aerosol particles. The *EF* is defined as the ratio of the number fraction of a given particle class in ICRs to that in ambient aerosol particles within the same size range ($0.6 \mu\text{m} \leq d_{va} \leq 4 \mu\text{m}$). For Pb-containing particles, the corresponding within-class Pb fraction is used. Values greater than 1 indicate enrichment in ICRs and are shown in bold; values less than 1 indicate depletion.

Particle type	Non-construction, MPC		Non-construction, CC		Construction, CC	
	Total	Pb-containing	Total	Pb-containing	Total	Pb-containing
SON	0.21	0	0.31	0	0.32*	2.24
BBA	0.21	0	0.31	0	0.23*	1.37*
MD-K/Al/Fe	4.15	1.33	6.23	0.66	2.89*	0.97*
MD-Ca	0	-	7.88	1.57	0.77*	2.17
Other	1.95	2.12	0	-	0.24*	1.36

* based on more than 50 ICRs



et al., 2022; Wolf et al., 2020b) can initiate deposition IN at tropopause temperatures, typically between approximately -55 and -80 °C, considerably lower than the -40 °C conditions investigated here. Kilchhofer et al. (2021) reported enhanced deposition IN activity only at -60 °C for raffinose particles after CC processing at -45 °C and $RH_w=105\%$, potentially due to porous particle formation (e.g., Adler et al., 2013) and subsequent pore condensation freezing. No IN activity enhancement was observed for levoglucosan particles across all MPC and CC conditions tested between -60 °C and -30 °C, as well as for raffinose particles above -60 °C because this was above the glassy transition temperature. Similarly, Wagner et al. (2017) observed negligible morphological changes and no enhancement in IN activity for α -pinene secondary organic aerosol particles after CC processing below -38 °C, whereas MPC processing mimicking a convective cloud system produced highly porous particles that acted as efficient immersion-freezing INPs. For BBA particles, laboratory studies (e.g., McCluskey et al., 2014; Umo et al., 2015; Grawe et al., 2016) and field observations (McCluskey et al., 2014; Córdoba et al., 2021) have shown that lofted mineral-containing soil or ash aerosol particles within wildfire plumes might exhibit high IN activity in immersion mode. Jahn et al. (2020) further reported that minerals originating from biomass in both BBA particles and bottom ash could initiate immersion freezing at temperatures as high as -13 °C. DeMott et al. (2009) reported that BBA particles undergoing glassy transitions between -60 °C and -45 °C still nucleate ice close to homogeneous freezing conditions, indicating their low deposition IN activity. Given the substantially higher experiment temperature in this study ($T = -40\text{ °C}$ and $RH_w = 90\%$), we expected the contribution of SON and BBA to ICRs under CC condition in this study should be negligible.

Moreover, no distinct SSA particle signatures were observed in either ambient aerosol particles or ICRs throughout the campaign, in contrast to previous observations at JFJ (Ebert et al., 2011; Worringer et al., 2015; Schmidt et al., 2017; Eriksson Hammer et al., 2018; Lacher et al., 2021). SSA particles are reported to be transported to JFJ in previous studies (Boose et al., 2016; Lacher et al., 2021). The absence of SSA particles in the present study may be explained by the prevailing transport patterns, as back-trajectory analysis indicates that the sampled air masses had a short residence time in the marine boundary layer and therefore limited opportunity to acquire SSA during transport (Figs. A3 and A4).

3.2.2 Natural and anthropogenic MD particles are the major ambient INP source

The contribution of MD particles to ambient aerosol particles with $0.1\text{ }\mu\text{m} < d_{va} \leq 4\text{ }\mu\text{m}$ increases from 2.5 % during the non-construction period to 7.5 % during the construction period (Fig. 4a and e), indicating a clear influence of anthropogenic MD particle emissions from local construction activities on ambient MD concentrations at JFJ. For ambient aerosol particles with $0.6\text{ }\mu\text{m} \leq d_{va} \leq 4\text{ }\mu\text{m}$, where MD particle presence is expected to be more pronounced, $\approx 12.1\%$ (208 of 1734) are classified as MD particles during the non-construction period (Fig. 4b), compared with 32.8 % (282 of 861) during the construction period (Fig. 4f). Across all size ranges and time periods (Fig. 4a, b, e, and f), ambient MD-K/Al/Fe particles are consistently 4 to 6 times more abundant than MD-Ca.

As shown in Fig. 4b-d and Table 1, natural MD (MD-K/Al/Fe and MD-Ca) aerosol particles with $0.6\text{ }\mu\text{m} \leq d_{va} \leq 4\text{ }\mu\text{m}$ during the non-construction period are enriched by a factor of ≈ 3.31 and 6.61 after MPC and CC processing in HINC-Auto, respectively. These values indicate clear trends but have low statistical significance due to the limited ICR sampling numbers during the non-construction period. During the construction period, when large amounts of anthropogenic MD particles are

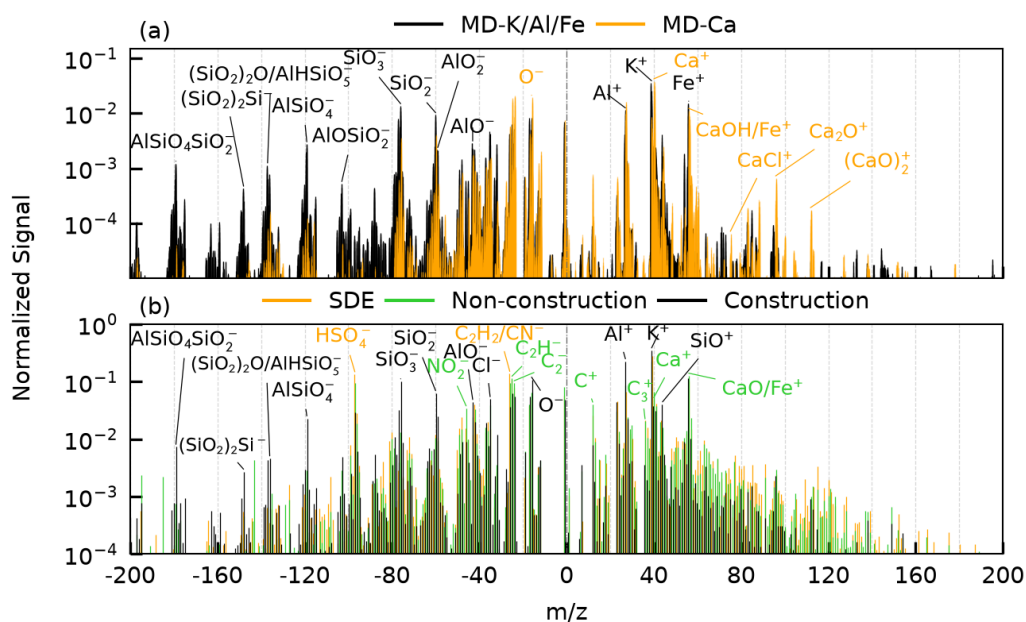


Figure 5. Normalized (a) PALMS-NG spectra of an MD-K/Al/Fe particle sampled on February 14 (during construction periods) and an MD-Ca particle sampled on February 3 (non-construction periods), (b) Average PALMS-NG spectra of MD particles during different periods. For visualization clarity, the MD-K/Al/Fe and MD-Ca spectra in panel (a), as well as the SDE and Construction MD spectra in panel (b), are displayed with m/z offsets of -0.2 and +0.2, respectively, to reduce overlap between the spectra.

released, they are enriched by a factor of ≈ 2.52 in ICRs collected after CC processing, with MD-K/Al/Fe enriched by a factor of ≈ 2.89 relative to ambient aerosol particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ (Table 1). These *EF*s are conservative and represent the lower limit, because ICRs with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ detected by PALMS-NG may originate from ambient aerosol particles in the $0.1 \mu\text{m} < d_{\text{va}} \leq 0.6 \mu\text{m}$ range. When compared to the full size range ambient aerosol particle size range ($0.1 \mu\text{m} < d_{\text{va}} \leq 4 \mu\text{m}$, Fig. 4a and e), the *EF* for MD particles exceeds 10 after both MPC and CC processing during both construction and non-construction periods. The consistent enrichment of MD particles downstream of the PCVI demonstrates that both natural and anthropogenic MD aerosol particles at JFJ act as effective INPs in both immersion freezing and deposition IN modes. Previous MPC IPR studies at JFJ reported similar enhancements for MD or metallic species, though the classification and *EF* magnitudes vary due to instrument differences (Mertes et al., 2007; Kamphus et al., 2010; Ebert et al., 2011; Worringer et al., 2015; Kupiszewski et al., 2016; Schmidt et al., 2017; Eriksen Hammer et al., 2018; Lacher et al., 2021). Comparable enhancement of MD species has also been observed for deposition ICRs at the Storm Peak Laboratory (Cziczo et al., 2003; DeMott et al., 2003; Richardson et al., 2007; Baustian et al., 2012).

K-feldspar exhibits higher immersion-freezing activity than many other mineral dust components, often nucleating ice at warmer temperatures in laboratory experiments (e.g., Atkinson et al., 2013; Augustin-Bauditz et al., 2014; Kaufmann et al.,



2016; Peckhaus et al., 2016; Welti et al., 2019), particularly due to active sites on freshly exposed surfaces (Kiselev et al., 2017; Keinert et al., 2022). However, Kaufmann et al. (2016) and Iwata and Matsuki (2018) showed that MD particles lacking K-feldspar could exhibit comparable IN activity. Holden et al. (2021) also demonstrated immersion freezing and deposition IN rely on distinct active sites, and effective deposition sites require size and/or pore geometry relevant for pore condensation and freezing. The ambient MD aerosol population at JFJ during this study is dominated by MD-K/Al/Fe across all size ranges and time periods, and consistently exhibit higher *EFs* compared to MD-Ca under MPC conditions during the non-construction period and under CC condition during the construction period, indicating generally higher IN activity in both immersion freezing and deposition IN modes. The lower deposition IN activity of MD-Ca is further supported by their slight depletion (*EF* = 0.77, Table 1) relative to ambient aerosol particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ during the construction period (Fig. 4f and g).

SON material coatings are known to suppress the IN activities of MD aerosol particles in both immersion freezing and deposition IN modes by altering particle surface properties (e.g., Gallavardin et al., 2008; Baustian et al., 2012). The small amounts of organics and sulfate for both MD types shown in Fig. 5a suggest minimal atmospheric aging. To address the role of biological material associated with MD on their IN activity (e.g., Prenni et al., 2009; Conen et al., 2011; O'Sullivan et al., 2014; O'Sullivan et al., 2015; Steinke et al., 2016; O'Sullivan et al., 2018), we attempted to quantify the biological signatures in MD ICR using the method proposed by Zawadowicz et al. (2017), which was developed based on prescribed laboratory biological species. However, the results were inconclusive due to limited ICR sample numbers. Additional field observations and biological mass spectra signal retrieval algorithm for field data are needed to assess the contribution of biological material to MD IN activity.

3.2.3 Frequent occurrence of Pb-containing aerosol particles in ICR

During the non-construction period, PALMS-NG detected 1658 Pb-containing ambient aerosol particles (6.1 % of 27153, hatched areas in Fig. 4a) in the size range $0.1 \mu\text{m} < d_{\text{va}} \leq 4 \mu\text{m}$. Among these, 1108 were classified as BBA and 251 as SON (66.7 % and 15.1 % of 1658, respectively). The overall Pb-containing aerosol particle fraction is consistent with the latest wintertime observations at JFJ, where up to 7 % of MPC IPRs contain Pb (Schmidt et al., 2017; Eriksen Hammer et al., 2018). For larger ambient aerosol particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$, 289 (16.7 % of 1734) particles contained Pb, including 156 (54.0 % of 289) BBA and 42 (14.5 % of 289) MD-K/Al/Fe (hatched areas in Fig. 4b). During the construction period, 806 (9.8 % of 8257, hatched areas in Fig. 4e) particles in the size range $0.1 \mu\text{m} < d_{\text{va}} \leq 4 \mu\text{m}$ exhibited Pb signature, and 218 (25.3 % of 861, hatched areas in Fig. 4f) particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ contained Pb.

Out of the 2248 ICR spectra collected, 572 (25.4 %) showed Pb signature across all periods (Fig. 4c, d, and g), among which 17.5 % were Pb-containing BBA ICRs and 69.4 % were MD-K/Al/Fe deposition ICRs collected during the construction period (Fig. 4g). Similarly high Pb occurrence in ICRs (more than 20 %) has been reported at Storm Peak Laboratory using comparable methods (Baustian et al., 2012). In contrast, Worringer et al. (2015) only observed rare appearance of Pb-containing MPC IPR at JFJ where the Pb appeared as internal mixtures.



Anthropogenic Pb commonly originates from general aviation gasoline in the form of tetraethyl lead $\text{Pb}(\text{C}_2\text{H}_5)_4$ (e.g.,
 385 Murphy et al., 2007), but can also be emitted from coal combustion, industry processes, as well as old building materials (e.g.,
 Shotyk and Roux, 2005). Cziczko et al. (2009) noted that the presence of Pb in ambient aerosol particles and MPC IPR is
 a strong indicator of anthropogenic influence. Emissions from aviation gas associated with general aviation aircraft near the
 station represent a plausible source. Previous laboratory studies have shown that Pb compounds are effective INPs (Schäfer,
 1954; Reischel, 1975) and trace amount of Pb doping could enhance the immersion freezing activity of MD particles (Cziczko
 390 et al., 2009). Welti et al. (2019) found a strong correlation between immersion freezing activity of K-feldspar and their trace Pb
 content. Under CC formation condition examined here, however, Pb does not appear to enhance the deposition IN activity of
 MD-K/Al/Fe. As shown in Fig. 4f and g and Table 1, the deposition IN activity of MD-K/Al/Fe particles seems to be unaffected
 by the presence Pb ($EF = 0.97$), as the relative proportions of Pb-containing MD-K/Al/Fe particles in ambient aerosol particles
 with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ (23.5 %, $\frac{6.4 \%}{27.2 \%}$) and ICRs (22.8 %, $\frac{17.9 \%}{78.5 \%}$) are similar, suggesting no enhancement effect.

395 Plants can also accumulate Pb from soil (e.g., Shotyk and Roux, 2005; Bi et al., 2018; Liu et al., 2023, and references
 therein), meaning that biomass burning can emit Pb-containing BBA particles. Such BBA particles might exhibit elevated IN
 activity in immersion mode (e.g., Jahn et al., 2020). Pb-containing SON, BBA, and MD-Ca particles appear to act as more
 effective deposition INPs under the CC formation condition in this study, because their fractions in ICRs are 1.37 to 2.24
 times larger than their corresponding fractions in ambient aerosol particles with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ (Table 1). Several
 400 previous MPC IPR observations at JFJ have reported similar enrichment of Pb-containing MPC IPRs relative to ambient
 aerosol particles (Cziczko et al., 2009; Kamphus et al., 2010; Ebert et al., 2011; Worringer et al., 2015; Schmidt et al., 2017;
 Eriksen Hammer et al., 2018).

3.3 Impact of meteorological conditions on ICR population

3.3.1 Sporadic meteoric (MT) aerosol particle occurrence from stratospheric air intrusions

405 MT aerosol particles are commonly observed during aircraft campaigns near the tropopause (e.g., Murphy et al., 2006; Schnei-
 der et al., 2021; Adachi et al., 2022a) and have also been identified in cirrus IPRs, indicating a potential link between strato-
 spheric aerosols and tropospheric cloud formation (Cziczko et al., 2004). PALMS-NG detected 50 ambient aerosol particles
 showing MT signatures, including abundant $m/z = 24 \text{ Mg}^+$ and $m/z = 56 \text{ Fe}^+$ ions with Mg/Fe ion ratios within the range
 typically reported for MT particles (e.g., Cziczko et al., 2001; Murphy et al., 2023) in the size range $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$
 410 (Fig. 6c) during the non-construction period, occurring mainly on the mornings of January 23, February 1, and February 5.
 These MT particles were most likely transported to the station via stratospheric intrusions. Their occurrence coincided with
 low RH_w , elevated O_3 , and enhanced PV, all of which are indicative of stratospheric influence (Fig. 6a and d, Fig. 7; Stohl
 et al., 2003; Cristofanelli et al., 2009; Trickl et al., 2016). JFJ is influenced by stratospheric intrusions $\approx 20 \%$ of the time (Cui
 et al., 2009; Bindschedler, 2025).

415 Laboratory studies have shown that the IN activity of metal oxides depends strongly on their cationic composition (e.g.,
 Souda et al., 2020; Chong et al., 2021; Schiffman et al., 2023). For example, Fe oxides are effective INPs via both immersion



Table 2. Number and percentage of PALMS-NG spectra for ambient aerosol particles and ICRs in the size range $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ when the station is in cloud ($RH_w \geq 90 \%$, Fig. 2b). The solid lines above February 8 and 11 are used to divide the MPC and CC processing during the non-construction (Non-cons.) and the CC processing during the construction periods (Cons.).

Classes	SON		BBA		MD		Others	
PALMS-NG	Amb.	ICR	Amb.	ICR	Amb.	ICR	Amb.	ICR
2024-01-22	0	0	1 (5.9 %)	0	12 (70.6 %)	0	4 (23.5 %)	0
2024-01-23	7 (7.4 %)	0	7 (7.4 %)	0	31 (33.0 %)	0	49 (52.1 %)	0
2024-01-24	17 (51.5 %)	0	7 (21.2 %)	0	1 (3.0 %)	0	8 (24.2 %)	0
2024-01-31	0	0	0	0	0	1 (100.0 %)	0	0
2024-02-01	3 (21.4 %)	0	2 (14.3 %)	0	4 (28.6 %)	0	5 (35.7 %)	1 (100.0 %)
2024-02-02	0	0	0	0	1 (100.0 %)	0	0	0
Non-cons. MPC	27 (17.0 %)	0	17 (10.7 %)	0	49 (30.8 %)	1 (50.0 %)	66 (41.5 %)	1 (50.0 %)
2024-02-08	35 (47.9 %)	0	21 (28.8 %)	0	6 (8.2 %)	0	11 (15.1 %)	0
2024-02-09	1 (16.7 %)	0	1 (16.7 %)	0	1 (16.7 %)	0	3 (50.0 %)	0
Non-cons. CC	36 (45.6 %)	0	22 (27.8 %)	0	7 (8.9 %)	0	14 (17.7 %)	0
2024-02-11	6 (35.3 %)	0	4 (23.5 %)	0	3 (17.6 %)	0	4 (23.5 %)	0
2024-02-12	16 (31.4 %)	2 (10.5 %)	21 (41.2 %)	6 (31.6 %)	1 (2.0 %)	8 (42.1 %)	13 (25.5 %)	3 (15.8 %)
2024-02-14	7 (16.3 %)	5 (9.6 %)	23 (53.5 %)	14 (26.9 %)	3 (7.0 %)	26 (50.0 %)	10 (23.3 %)	7 (13.5 %)
Cons. CC	29 (26.1 %)	7 (9.9 %)	48 (43.2 %)	20 (28.2 %)	7 (6.3 %)	34 (47.9 %)	27 (24.3 %)	10 (14.1 %)

freezing and deposition IN modes (Saunders et al., 2010; Hiranuma et al., 2014; Chong et al., 2021; Schiffman et al., 2023), whereas MgO exhibits low immersion freezing activity due to weak surface water binding and limited ice growth potential (Souda et al., 2020). In our observations, MT aerosol particles were absent in ICRs during the non-construction period, likely reflecting either their low IN activity under both MPC and CC formation conditions or low abundance.

During the construction period, PALMS-NG identified 10 ambient aerosol particles and 6 ICRs with $0.6 \mu\text{m} \leq d_{\text{va}} \leq 4 \mu\text{m}$ as MT particles. All 6 MT ICRs, along with 2 ambient MT aerosol particles, were observed on February 13 when RH_w was $35.0 \% \pm 3.4 \%$ and O_3 reached 53.1 ± 1.8 ppm indicative of stratospheric intrusion (Fig. 6a). On this day, MT particles showed $EF \approx 0.46$ (0.4 % of 1481 ICRs and 0.9 % of 228 ambient aerosol particles), which may indicate comparatively low deposition IN activity under the conditions examined in this study. However, due to the potential interference from construction activities and the limited sample number, these results should be treated qualitatively.

3.3.2 Uncertainty in the impact of natural cloud processing on the IN activity of ambient aerosol particles

As described in Sect. 3.1, the occurrence of supercooled liquid clouds or MPCs at JFJ during the campaign was strongly associated with BLI. When the station was in cloud, the dried sample air from the total inlet contained interstitial aerosol

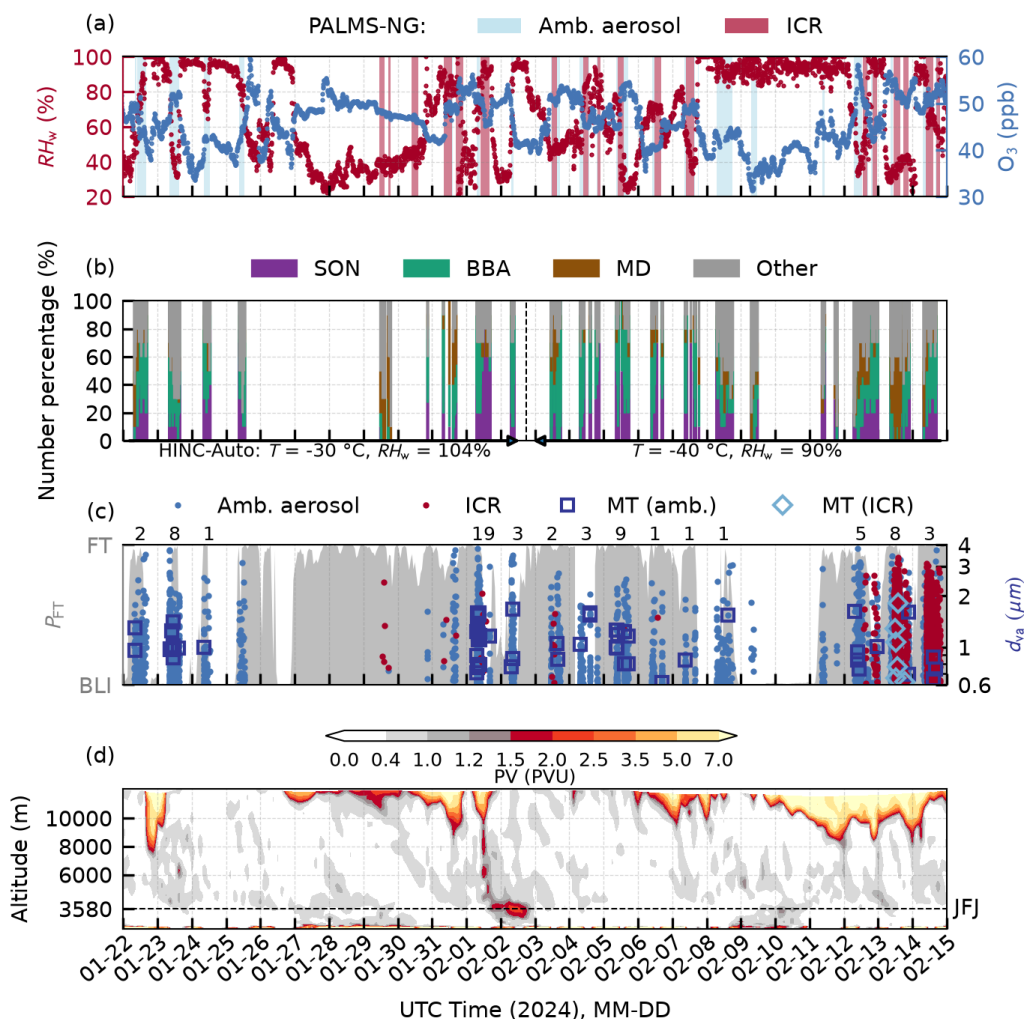


Figure 6. Time series of (a) RH_w and O_3 , (b) number percentage of particle classes measured by PALMS-NG, (c) MT aerosol particle occurrence during the campaign, and (d) PV at JFJ derived from ERA5. Numbers on the top of panel (c) denote daily counts of MT particles in the size range $0.6 \mu\text{m} \leq d_{va} \leq 4 \mu\text{m}$. The dashed line in panel (d) indicates the altitude of JFJ. Episodes of potential stratospheric intrusion are evident on January 23 and 31, as well as February 1, 7, 12, and 13.

430 particles and cloud hydrometeor residuals that have undergone natural cloud processing. These conditions provide a natural laboratory for investigating the IN activity of aerosol particles after experiencing in-cloud scavenging, aqueous chemistry, as well as dry-freezing cycles, all of which can alter aerosol particle morphology and chemical composition (e.g., Yao et al., 2021; Huang et al., 2025).

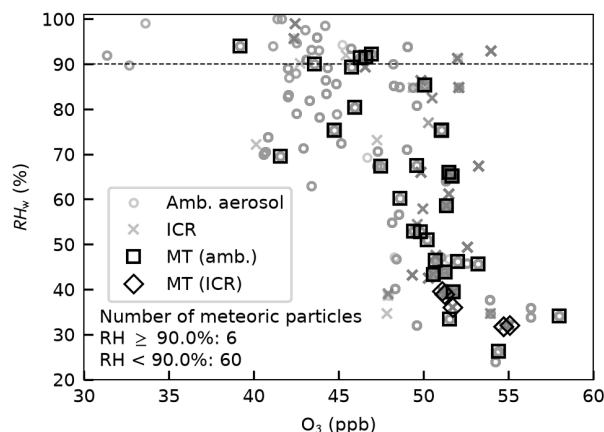


Figure 7. RH_w as a function of O_3 for particles with $0.6 \mu m \leq d_{va} \leq 4 \mu m$ detected by PALMS-NG. Ambient aerosol particles are shown as circles and ICRs as crosses. MT particles are highlighted by squares in ambient aerosol samples and by diamonds in ICRs.

During the construction period, the overall fraction of MD particles in the ICRs is 82.8 % with an EF of 2.52 (Sect. 3.2.2 and Fig. 4f and g), whereas the fraction drops to below 50 % but the EF increase to 7.59 (Table 2, $\frac{47.9\%}{6.3\%}$) when the station is in cloud. The increased EF suggests an enhancement of deposition IN activity of MD aerosol particles sampled during in-cloud periods. This observation is in contrast to the results from Kilchhofer et al. (2021), who reported decreased deposition IN activity below $-45^\circ C$ for coal fly ash particles after MPC processing at $-30^\circ C$ and $RH_w = 108\%$. This was attributed to hydration of the hygroscopic $CaSO_4$ in the MPC processing. As shown in Fig. 4e and f, this may be related to dominance of the relatively fresh MD-K/Al/Fe type of the MD population during the construction period in this study, which may have experienced less atmospheric ageing prior to sampling and therefore is less hygroscopic compared to MD-Ca (Fig. 5b).

The EF for BBA particles increases significantly from 0.23 to 0.65, whereas that for SON aerosol particles drops only slightly from 0.32 to 0.38. The drop in the depletion factor suggests enhanced deposition IN activity after natural cloud processing for BBA particles. However, further investigation is needed to establish the impact of MPC processing on the IN activity of BBA particles, as no ICRs associated with BBA were directly observed in the present study. Wagner et al. (2017) observed highly porous organic particles production after MPC processing, but such morphological change did not impose significant impact on the deposition IN activity of these organic particles. The observed deposition IN activity enhancement after natural cloud processing for BBA particles here might be due to the associated Pb content as discussed in Sect. 3.2.3 or morphology change of the organic content associated to BBA particles after a dry-freezing cycle.

During the non-construction period (Table 2), ICR occurrence was rare under both MPC and CC conditions when the station was in cloud, primarily due to the low N_{CPC} during the PALMS-NG operation periods (Sect. 3.1 and Fig. 3b). Previous studies of MPC IPR at JFJ attributed the appearance of soluble IPRs to ice particles produced from secondary ice production processes or cloud droplet residuals, which occupied low immersion freezing and deposition IN activities as discussed in Sect. 3.2.1.



4 Conclusions

455 This study presents the wintertime online chemical composition characterization of ICRs at JFJ and identifies the aerosol classes responsible for initiating IN under conditions representative of MPC ($T = -30\text{ }^{\circ}\text{C}$, $RH_w = 104\%$) and CC ($T = -40\text{ }^{\circ}\text{C}$, $RH_w = 90\%$).

Across all investigated conditions, MD emerged as the dominant INP despite its low ambient number concentration. Two subclasses, namely MD-K/Al/Fe and MD-Ca, were distinguished, with MD-K/Al/Fe consistently showing higher *EF*s and therefore higher IN activity in both immersion freezing and deposition IN modes. During periods influenced by nearby construction activities, anthropogenic MD-K/Al/Fe particles contributed substantially to the deposition INP population under CC conditions, highlighting the sensitivity of INP observations to local emissions. These results underscore that anthropogenic activities can impose direct impact on ambient INP observation in remote environments. The occurrence of MD ICRs outside the construction period further suggests the presence of additional regional or long-range transported MD sources.

465 In contrast, SON and BBA particles, which dominated the ambient aerosol particle population, were strongly depleted in ICRs under both MPC and CC conditions. Their low occurrence in ICRs indicates limited immersion freezing activity at $T = -30\text{ }^{\circ}\text{C}$, $RH_w = 104\%$ and limited deposition IN activity at $T = -40\text{ }^{\circ}\text{C}$, $RH_w = 90\%$, in agreement with previous field and laboratory observation.

Pb-containing particles occurred frequently in ICRs, particularly among BBA particles. More than half of the detected BBA ICRs contained Pb signatures potentially associated with residential biomass-burning emissions, suggesting that Pb may enhance the IN activity of specific particle populations. In contrast, no clear enhancement was observed for MD-K/Al/Fe particles, for which similar fractions of ambient particles and ICRs contained Pb.

Sporadic MT particles were detected during periods characterized by stratospheric intrusions. Although MT particles were only rarely observed in ICRs, their occurrence demonstrates that meteoric material can be transported to JFJ and suggests a potential pathway linking stratospheric aerosols to tropospheric ice formation. Additional observations are required to determine whether such particles contribute significantly to atmospheric INP populations, particularly in the mid-troposphere, where their concentrations are low. An emerging source of anthropogenic aerosol and INPs to consider in the upper troposphere in the future is debris from space rocket re-entry, as these particles are directly emitted in the upper troposphere through ablation and fragmentation of heat-shield materials and rocket body components during descent.

480 Observations during in-cloud periods suggest that natural cloud processing may alter the deposition IN activity of selected aerosol particle classes following a dry-freezing cycle, particularly MD-K/Al/Fe and BBA. However, the limited number of in-cloud ICRs, likely due to efficient in-cloud scavenging and/or IN, limits a robust assessment of cloud-processing effects.

Overall, our results demonstrate that MD, from both natural and anthropogenic sources, dominated the wintertime INP populations at JFJ, whereas other aerosol classes played comparatively minor roles in atmospheric IN under the investigated conditions. These findings highlight the need to account for local MD emissions, particle composition, and aging processes when assessing INP variability. Future field observations over longer periods and additional seasons will be essential for



quantifying the contributions of the biogenic material associated with MD, long-range transported material, natural cloud processing, Pb-containing particles, and episodic stratospheric aerosol sources to ambient INP populations.

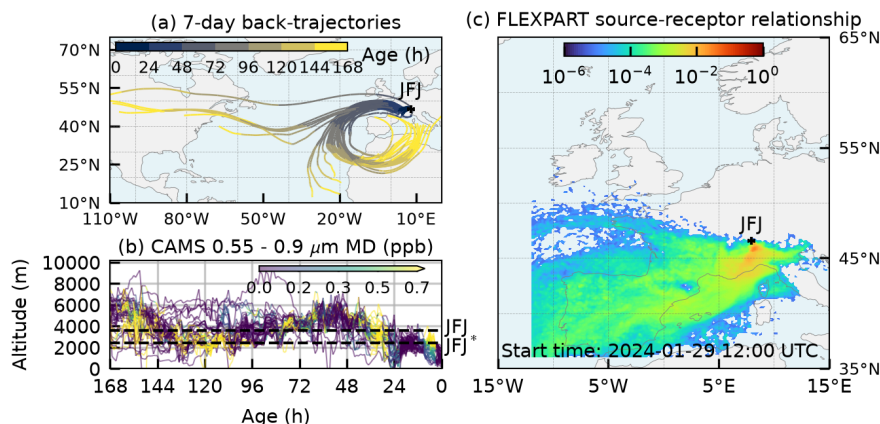


Figure A1. (a) 7-day back trajectories, (b) CAMS 0.55 - 0.90 μm MD (ppb), and (c) source-receptor relationship for January 29, showing that the air mass and associated aerosol population arriving at JFJ passed through the northwestern Sahara and Spain. The actual altitude and model altitude of JFJ are denoted as JFJ and JFJ* in panel b.

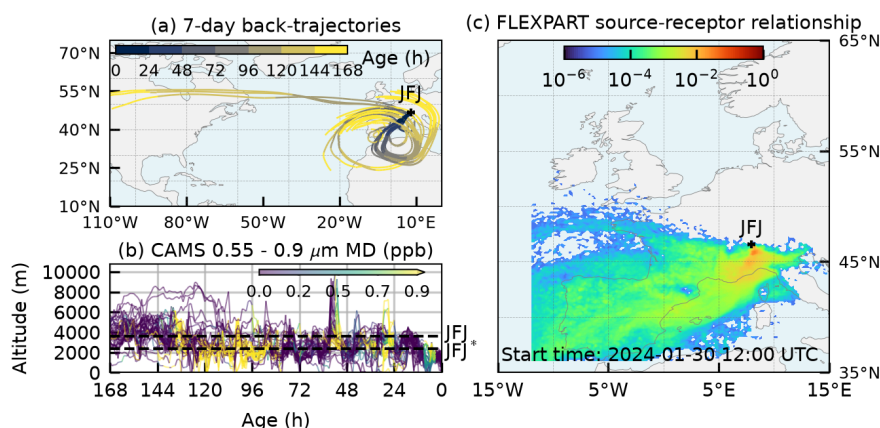


Figure A2. (a) 7-day back trajectories, (b) CAMS 0.55 - 0.90 μm MD (ppb), and (c) source-receptor relationship for January 30, showing that the air mass and associated aerosol population arriving at JFJ passed through the northwestern Sahara and Spain.

Data availability. The data used to produce the figures in this manuscript will be made available through a persistent DOI upon acceptance of the manuscript. Meteorological data at Jungfraujoch is available at <https://opendatadocs.meteoswiss.ch/a-data-groundbased>. ERA5 reanalysis data are publicly available from the Copernicus Climate Data Store (<https://cds.climate.copernicus.eu/>). CAMS reanalysis data are publicly available from the Copernicus Atmosphere Data Store (<https://ads.atmosphere.copernicus.eu/>).

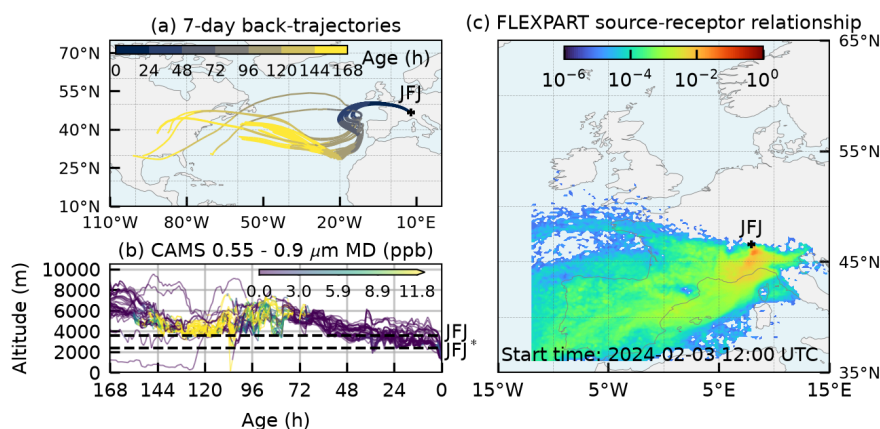


Figure A3. (a) 7-day back trajectories, (b) CAMS 0.55 - 0.90 μm MD (ppb), and (c) source-receptor relationship for February 3, showing that the air mass and associated aerosol population arriving at JFJ passed through the Atlantic Ocean.

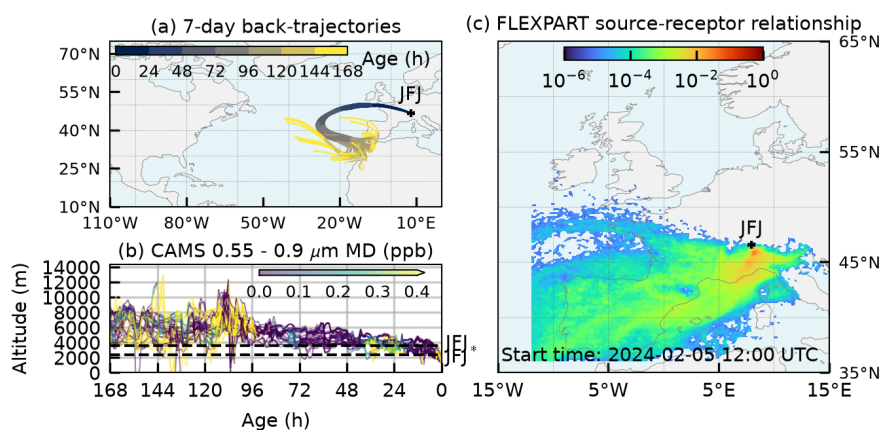


Figure A4. (a) 7-day back trajectories, (b) CAMS 0.55 - 0.90 μm MD (ppb), and (c) source-receptor relationship for February 5, showing that the air mass and associated aerosol population arriving at JFJ passed through the Atlantic Ocean.

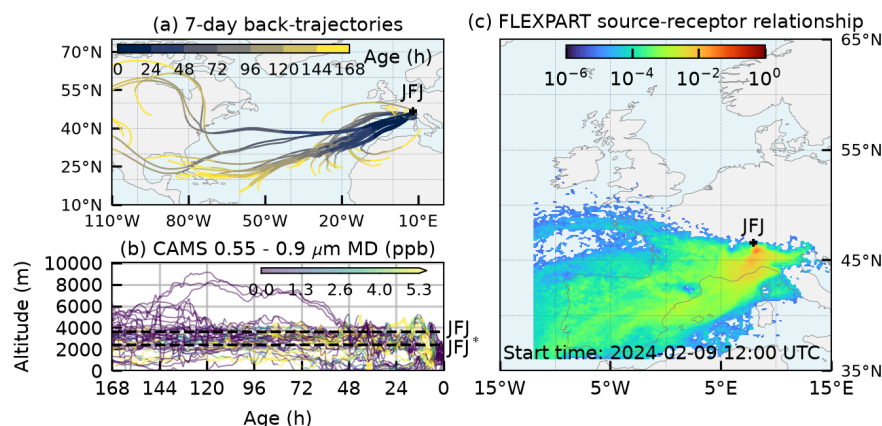


Figure A5. (a) 7-day back trajectories, (b) CAMS 0.55 - 0.90 μm MD (ppb), and (c) source-receptor relationship for February 9, showing that the air mass and associated aerosol population arriving at JFJ passed through the Po Valley.

Appendix A: Lagrangian back trajectories and aerosol source-receptor relationship

Author contributions. CZ, DJC, and ZAK conceptualized the experiment. CZ, XS, JJ conducted the campaign with support from LL, NN, BB, and RM. CZ and XS analyzed and visualized the data. XS analyzed the PALMS-NG data. NN, BB, RM, and MGB provided the aerosol data. MSt provided the reactive trace gas and meteorology data. FC provided the ²²²Rn data. VS and MSp configured LAGRANTO and performed the analysis. SH provided FLEXPART data. MCC provided the list of SDE alerts. CZ, XS, JJ, DJC, and ZAK prepared the article with input from all co-authors. ZAK and DJC supervised the study.

Competing interests. Ottmar Möhler is a member of the editorial board of Atmospheric Chemistry and Physics and a co-author of this paper.

Disclaimer. During the preparation of this work, CZ used Microsoft Copilot to assist with text rephrasing and with code corrections for data analysis and visualization. All tool-assisted content was reviewed, edited, and validated by CZ.

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ERIC) provided by the Swiss State Secretariat for Education and Research and Innovation (SERI). Additional observations for the purpose of this study received financial support from the European Union's Horizon 2020 Research and Innovation Programme under Grant Agreement N° 101008004 (ATMO ACCESS; trans-national access scheme). The radon observations are supported by the Swiss National Science Foundation (SNSF) as a contribution to the pan-European Integrated Carbon Observation System, ICOS (SNSF grant 20FI20_198227).

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