



Drivers of ice-nucleating particle variability at urban and high-mountain environments of the Western Mediterranean

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Abstract.

This study presents a phenomenological analysis of immersion freezing ice-nucleating particles (INPs) at two different environments, urban (UGR) and high-mountain (SNS) stations, in Southern Spain. Measurements were conducted from June 2024 until June 2026, including INP concentration spectra and aerosol properties. The INP concentrations at UGR and SNS were consistent with previous measurements in the Mediterranean, showing a seasonal pattern. Spring shows the highest INP concentration values at high temperatures, which is likely related to the presence of biological particles. In contrast, during summer and autumn INP concentration values were higher at lower temperatures, which can be linked to the presence of Saharan dust. A correlation analysis between INP concentrations and aerosol optical properties (AOPs) revealed that at UGR there is negative correlation with the single scattering albedo (SSA), whereas for SNS there is positive correlation between INP concentrations and aerosol load proxies, such as the scattering and absorption coefficients. After calculating permutation feature importances between the INP concentrations and the AOPs, we developed parameterizations to predict INP concentrations. At UGR the parameterization is based on SSA, whereas at SNS the parameterization is based on aerosol scattering coefficient. For both parameterizations, 97 % percent of the data fall within a factor 10, and around 90 % of the data is within a factor 5, which reveals a good agreement between predicted and observed INP concentrations. These prediction schemes based on AOPs open new possibilities to extend INP estimations to observatories that routinely measure AOPs, extending the spatial and temporal coverage of INPs.

1 Introduction

Atmospheric aerosol particles play a key role in cloud formation and evolution through aerosol-cloud interactions (ACI), which remain one of the largest sources of uncertainty in climate models (Forster et al., 2021). A subset of aerosol particles can act as ice nucleating particles (INPs), triggering the freezing of supercooled water droplets at temperatures above the homogeneous freezing temperature (-38°C). Heterogeneous freezing can occur through several mechanisms, among which immersion freezing is considered the dominant pathway in mixed-phase clouds (Kanji et al., 2017). Although INP concentrations are several orders of magnitude lower than aerosol particle concentrations, they play a crucial role in cloud microphysics, as the ice phase



is responsible for most precipitation over land from mixed-phase clouds (Heymsfield et al., 2020; Mülmenstädt et al., 2015).
 25 Moreover, aerosol properties influence cloud properties such as lifetime and albedo (Haywood and Boucher, 2000). Therefore, understanding how different aerosol types govern atmospheric INP activation is essential to improve the representation of aerosol-cloud interactions in climate models.

Among natural sources, atmospheric mineral dust particles are considered the most important contributor to the INP population (Hoose and Möhler, 2012), producing high INP concentrations generally at temperatures below -15 °C (DeMott et al.,
 30 2015). On the other hand, biological aerosol particles dominate the INP population at relatively warm temperatures (Cornwell et al., 2023; Huang et al., 2021). The Mediterranean is a unique region for the study of biological (Tang et al., 2022) and dust INPs (Beall et al., 2022), especially considering the proximity to the Sahara desert, which is responsible for almost 50 % of the global dust emissions (Kok et al., 2017). However, most INP studies in recent years have been developed in the Eastern Mediterranean region (Beall et al., 2022; Gao et al., 2024; Tang et al., 2022; Tarn et al., 2020), with few studies performed in
 35 the Western Mediterranean (Bras et al., 2024), thus limiting our ability to assess the representativeness of existing parameterizations and to understand the factors controlling INP concentrations in this region. This knowledge gap is particularly relevant given the complex aerosol environment of the Mediterranean, where mineral dust transported from North Africa interacts with marine, biogenic, and anthropogenic aerosols. Such interactions, together with the strong spatial and temporal variability of aerosol sources and atmospheric transport, may substantially influence the abundance and characteristics of atmospheric INPs.
 40 Expanding observational datasets in the Western Mediterranean is therefore essential to improve our understanding of the regional sources and variability of INPs and their potential implications for cloud processes.

In a global context, many INP studies have taken place in pristine environments, such as the Arctic region (Schneider et al., 2021; Wex et al., 2019) or high-mountain areas (Brunner et al., 2022; Conen et al., 2015; Lacher et al., 2021), as well as in urban locations (Mendoza-Téllez et al., 2026; Tobo et al., 2020). In particular, remote and pristine environments
 45 provide an ideal framework for isolating the ice nucleation properties of natural aerosols because they are less affected by local anthropogenic emissions. On the other hand, urban stations allow for the characterization of the INP ability of black carbon (BC) and other anthropogenic pollutants (Bi et al., 2019). However, most studies only span a few months of measurements, with only some sites covering years of INP properties (see Tables S1 and S2 for a detailed summary of high-mountain and urban INP measurements). Moreover, long-term observations combining a high-mountain site with a nearby urban site are still lacking.
 50 Such a tandem approach provides a unique opportunity to investigate how the interactions between aerosol particles from different sources influence the INP concentrations under comparable synoptic conditions. Therefore, long-term measurements of INP concentrations at both urban and high-mountain environments in the Western Mediterranean become highly relevant in this context.

Reliable INP parameterizations are crucial to improve the representation of INP concentrations in global climate models.
 55 Early parameterizations relied exclusively on the temperature dependence of the INP concentration (Meyers et al., 1992), although more recent parameterizations incorporate the relation between INP concentrations and aerosol properties. In particular, several parameterizations incorporate aerosol particle concentrations, particularly coarse particle concentrations, to account for the abundance of potential INPs (DeMott and Prenni, 2010; DeMott et al., 2015). In particular, DeMott and Prenni (2010)



demonstrated the importance of including aerosol particle concentrations, predicting 62 % of observed INP concentrations within a factor of 2, compared with only 16 % using the temperature-only parameterization of Meyers et al. (1992). However, in-situ measurements of coarse-mode aerosol particle concentrations are not widely available globally. Consequently, estimates of INP concentrations based on aerosol particle concentration measurements remain limited by the sparse spatial and temporal coverage of aerosol coarse measurements. In this sense, aerosol optical properties (AOPs) can serve as a proxy for aerosol load and microphysical properties, and therefore be used to develop INP parameterizations, since they are routinely measured in many atmospheric stations. Recently, AOPs have been successfully used to develop parameterizations of cloud condensation nuclei (CCN) concentrations (Shen et al., 2019; Zabala et al., 2026), providing satisfactory results. Therefore, the development of INP parameterizations based on aerosol optical properties would allow to extend the INP concentration estimates to locations where aerosol optical properties are available, improving the spatial coverage of INPs.

In this work we present simultaneous long-term observations of INP concentrations at urban and high-mountain stations that belong to the Andalusian Global Observatory of the Atmosphere (AGORA), located in Southern Spain. Measurements took place from June 2024 until June 2026, covering two years of data. Additional optical and microphysical aerosol measurements were also conducted, and used to develop different parameterizations to predict INP concentrations at both stations, allowing for a comprehensive study of INPs in the Western Mediterranean.

2 Experimental sites and instrumentation

2.1 Experimental sites

The experimental measurements used in this study were carried out at two stations that belong to the Andalusian Global Observatory of the Atmosphere (<https://atmosphere.ugr.es/en/about/presentation/agora>, last access: 19/05/2026), located in Southern Spain, which is part of the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS). The first site is an urban background station (UGR, 37.18° N, 3.58° W; 680 m a.s.l.) located in the city of Granada. The main local source of aerosol particles at the UGR station is road traffic (Titos et al., 2014, 2017), with some influence of biomass burning aerosol during wintertime (Casquero-Vera et al., 2021; Titos et al., 2017). The orographic situation of the city of Granada favors air stagnation, which causes accumulation of pollutants (Patrón et al., 2017). The second station is a high-mountain station located in the Mojón del Trigo peak, in the north slope of the Sierra Nevada National Park (SNS, 37.10° N, 3.39° W; 2610 m a.s.l.). The station is surrounded by a wide variety of plant communities (Cariñanos et al., 2025), which contribute to the aerosol population in the site. Since both stations are separated 20 km in horizontal distance and the mountain range contains various valleys that can channel the air upwards, the advection of pollutants (Rejano et al., 2021) and pollen particles (Cariñanos et al., 2025) from the city of Granada is common, especially during the spring and summer seasons when the station is more frequently within the planetary boundary layer (PBL). Additionally, the two stations are located around 200 km away from the African continent, so long-range transported dust from the Sahara Desert frequently reaches the stations (Bazo et al., 2025; Cazorla et al., 2017; Lyamani et al., 2010), especially during the summer season.



Aerosol particles were sampled using a total inlet that consists of a stainless-steel tube that is 5 m long and has a diameter of 20 cm (Lyamani et al., 2008). Several pipes are located inside the tube for splitting the aerosol flow into the different instruments. The final connection to the instruments' inlets is done with conductive tubing avoiding bends. All measurements were performed at ambient conditions, without the use of aerosol dryers. Measurements are reported at standard pressure and temperature (0 °C and 1013.25 hPa).

2.2 Instrumentation

The main instrument used in this study is the GRANada Ice Nuclei Spectrometer (Bazo et al., 2026), a droplet freezing assay that uses droplet volumes of 100 µL and a cooling rate of 1 K min⁻¹ for obtaining the INP concentration. Further information of GRAINS' operation can be found in Bazo et al. (2026). For the sampling of filters needed to derive the INP concentration, two different devices have been used. The first one is the GRANada Sampling System (GRASS) an automatic low volume sampler that works at 5 L min⁻¹ and that is connected to the station main inlet. The second ones are high-volume samplers (CAV-A/MSb for SNS and CAV-A/Mb for UGR), which sample aerosol particles under a PM10 inlet at a flowrate of 500 L min⁻¹. At UGR and SNS the high volume samplers were used for INP analysis until December 2024 and May 2025, respectively; whereas after that GRASS was used as the main sampler for INPs in each station.

For the characterization of the aerosol optical properties, several instruments were used. An integrating nephelometer (TSI 3563), which operated at a flowrate of 15 L min⁻¹, measured the aerosol particle light scattering coefficient at 450, 550 and 700 nm between 7 and 170°. Measurements were corrected to the entire angular range with the correction proposed by Anderson and Ogren (1998). In addition, two multiwavelength aethalometers (AE33 and AE36s, Magee Scientific) were used to determine the aerosol light absorption coefficient and the equivalent black carbon (eBC) concentration at seven or nine different wavelengths, respectively. In particular, AE33 measured until April 2026 at UGR and until May 2026 at SNS, and after that AE36s was installed. Both instruments operated at a flowrate of 4 L min⁻¹ and the absorption measurements were corrected with the ones measured at 637 nm by the Multiangle Absorption Photometer (MAAP 5012, Thermo Fisher).

Lastly, the aerodynamic particle sizer (APS, TSI 3321) spectrometer was used to measure the aerosol size distribution of the coarse mode particles, which are more relevant within the INP population. The APS works at a flowrate of 5 L min⁻¹ and is able to detect particles from 0.5 to 20 µm in aerodynamic diameter. More information about the measurement period of each instrument at both stations can be found in Figures S1 and S2.

3 Methodology

The measurement period in this study covered 2 years, from 10th June 2024 until 7th June 2026. Polycarbonate and quartz filters were sampled every four days for 24 hours at both stations. Since GRASS was not available at the beginning of the measurement period, quartz filters were used for INP analysis following the Quartz punch washed method described in Bazo et al. (2026), who performed an evaluation of the two filters for INP concentration calculation obtaining comparable results. In particular, at UGR quartz filters were sampled until December 2024, and from January 2025 until June 2026, 47 mm polycarbonate filters



were used (Whatman®, 0.2 µm pore size). At SNS, we sampled quartz filters until May 2025 and the same polycarbonate filters were used onwards. At both stations, field blanks were taken approximately every month (see Figure S3).

125 For the INP analysis in the laboratory, the filter (or a 1 cm punch of the filter when sampling quartz filters) was washed in 10 mL of ultrapure filtered (0.1 µm syringe filter) water and it was agitated at 60 rpm for 20 minutes in a rotating agitator in order to extract the particles. The suspension was pipetted (100 µL droplets) into two PCR trays and they were introduced in GRAINS, with a cooling rate of 1 K min⁻¹. Then, the frozen fraction (FF(T)) was obtained as:

$$FF(T) = \frac{N_f(T)}{N_0}, \quad (1)$$

130 where N_0 is the total number of wells filled with the sample suspension and $N_f(T)$ is the number of wells with a frozen sample at temperature T (Vali, 1971). Then, the INP concentration was calculated as:

$$C_{INP} = -\frac{1}{V_{drop}} \frac{V_{suspension}}{V_{air}} \ln(1 - FF(T)), \quad (2)$$

where $V_{suspension}$ is the volume of water used to wash the filter and V_{air} is the proportional volume of air at standard conditions (0 °C and 1013.25 hPa) that passed through the filter. The INP concentration data were corrected by the background from the freezing spectra of the filtered ultrapure water used to make the suspensions. Taking into account the total surface area of aerosol particles S_{tot} (see Section 3.2 for details on S_{tot} calculation), the number of ice-nucleation-active sites (INAS) density (n_s) was obtained:

$$n_s(T) = \frac{C_{INP}}{S_{tot}}. \quad (3)$$

Starting in January 2025, 10- and 100-fold dilutions were used in the analysis of the filters from both stations to extend the
 140 INP concentration range towards lower temperatures.

3.1 Optical-derived properties

Optical properties were measured continuously at both stations, with a time resolution of 1 min. Several intensive properties were derived from the measured optical properties in order to classify different aerosol particles. In particular, the scattering and absorption coefficient measured by the integrating nephelometer and the AE33/36s, respectively, have been used to calculate
 145 the scattering Ångström exponent (SAE) and the absorption Ångström exponent (AAE):

$$SAE_{\lambda_1-\lambda_2} = -\frac{\ln\left(\frac{\sigma_{sp}(\lambda_1)}{\sigma_{sp}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)}, \quad (4)$$

$$AAE_{\lambda_1-\lambda_2} = -\frac{\ln\left(\frac{\sigma_{ap}(\lambda_1)}{\sigma_{ap}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)}. \quad (5)$$



The wavelengths used to calculate SAE were 450 and 700 nm, whereas for AAE we have used 370 and 660 nm. Additionally,
 150 we have computed the single scattering albedo (SSA) at 550 nm of the aerosol particles by:

$$SSA(\lambda) = \frac{\sigma_{sp}(\lambda)}{\sigma_{sp}(\lambda) + \sigma_{ap}(\lambda)}, \quad (6)$$

where σ_{ap} was calculated at 550 nm by means of the Ångström law.

3.2 Total surface area of aerosol particles

The total surface area of aerosol particles larger than 500 nm (S_{tot}) can be obtained from the size distributions measured by
 155 the APS. For this, aerodynamic diameter is first converted to a volume equivalent geometric diameter by choosing a particle
 density and a dynamic shape factor of the particles. Here, we have assumed spherical particles with a density of 1 g cm^{-3} .
 Then, the surface size distribution is obtained from the particle number size distribution and is integrated over the entire size
 range to obtain S_{tot} .

During the 2-years measurement period, the APS measurements have several gaps at both stations due to malfunction of
 160 the instrument (mostly problems with the laser in both APS units). To enhance the data coverage during those periods, and
 be able to calculate the INAS density, we have calculated S_{tot} using aerosol scattering measurements (Pantoya et al., 2025).
 Figure S4 shows the relationship between the total surface area and the scattering coefficient, colored by the SAE, during the
 co-located period of APS and nephelometer at UGR and SNS. As observed in Figure S4, the trend between both variables is
 linear but highly dependent on the SAE. Therefore, to predict S_{tot} from scattering measurements it is necessary to account for
 165 the effect of the size of the particles in the relationship between the scattering and the total surface area. We have applied a
 multi-parametric model based on σ_{sp} (450 nm) and the SAE to estimate S_{tot} for those periods when APS measurements were
 not available. To obtain this relationship we have randomly selected a 70 % of the available database for coincident optical and
 size distribution measurements, and then we have evaluated the model with the remaining 30 % of the database. In particular,
 we have applied a multi-parametric fit with the following equation:

$$170 \quad S_{tot} = a \cdot \sigma_{scat}(450 \text{ nm}) + b \cdot (\sigma_{scat}(450 \text{ nm}) \cdot \text{SAE}) + c \quad (7)$$

where the coefficients obtained for each station are shown in Table 1. In general, the coefficients obtained in both models are
 very similar, although there are slight differences in the coefficient related to σ_{sp} (450 nm).

Table 1. Fit parameters of Equation 7 for the data at UGR and SNS.

Station	$a \text{ } (\mu\text{m}^2 \text{ cm}^{-3} \text{ Mm})$	$b \text{ } (\mu\text{m}^2 \text{ cm}^{-3} \text{ Mm})$	$c \text{ } (\mu\text{m}^2 \text{ cm}^{-3})$	r^2
UGR	1.223 ± 0.022	-0.599 ± 0.015	4.7 ± 1.0	0.93
SNS	1.745 ± 0.075	-0.695 ± 0.051	4.7 ± 2.1	0.81

To evaluate the goodness of the reconstructed values, we have calculated the corresponding INAS densities with the measured
 values of the total surface of particles and the ones predicted by the model. Figure 1 shows the n_s data obtained with each



method, as well as reference lines corresponding to a 1:1 relationship and factors of 2, 5 and 10. Previous studies have shown that when performing intercomparisons of different instruments for the determination of INP, differences can be up to one order of magnitude (DeMott et al., 2025). Also, Lacher et al. (2024) proposed that there was agreement in INP concentrations if measurements were within factors of 2 and 5, which make the measurements suitable to derive model-relevant INP data. Therefore, we have selected these factors to evaluate the level of agreement of n_s derived from the optical properties.

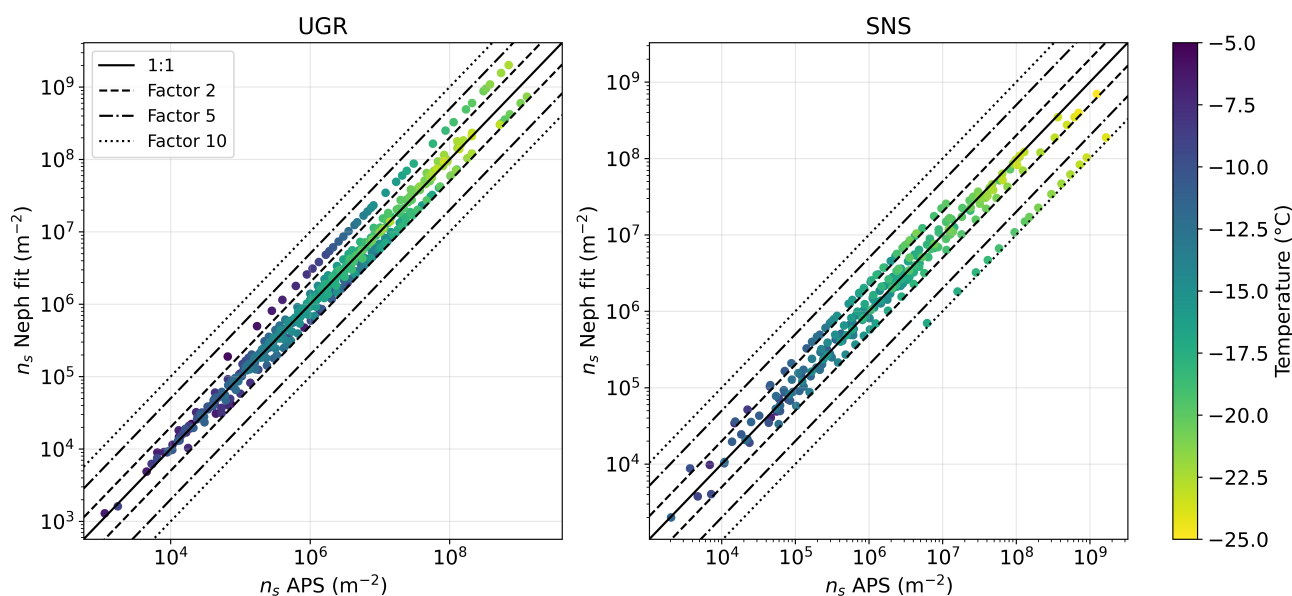


Figure 1. n_s obtained with the surface from the nephelometer model compared to the one obtained from APS measurements for UGR (left) and SNS (right).

In general, Figure 1 shows a very good agreement, with the majority of the data falling near the 1:1 line. In particular, for UGR, 93 % of the data fall inside the region delimited by a factor 2, and 100 % of the data are within factors 5 and 10. For SNS, 75 % of the data is within a comparison factor of 2, whereas 94 % of the data falls inside the region delimited by a factor 5. Also, for SNS all data points (100 %) fall inside a comparison factor of 10. These results show consistency between the data obtained with the experimental total aerosol surface and the one derived with the model based on optical properties. Therefore, this opens new possibilities for deriving INAS densities whenever size distribution measurements are not available for a long period of time. However, since the results of the model are clearly station-dependent, a first comparison between the surface of aerosol particles and the scattering coefficients should be performed.

4 Results and Discussion

4.1 Overview of INPs at UGR and SNS

Figure 2 shows boxplots of the INP concentration spectra, binned to 0.5 °C, at UGR and SNS for the whole measurement period. INP concentrations are reported from -5 °C to -24.5 °C at UGR, and from -5 °C to -26 °C at SNS. In general, at UGR there are higher INP concentrations than at SNS. This feature becomes more evident around the middle of the temperature range, whereas at the bounds of the temperature range the INP concentrations at both stations are more similar. Figure 2 also shows previous measurements of INP concentrations from the Eastern Mediterranean region (Beall et al., 2022; Gao et al., 2024; Tang et al., 2022; Tarn et al., 2020), as there are no studies reporting INP concentrations in the Western Mediterranean. As observed in Figure 2, INP data from UGR and SNS fall in the region of measurements in the Mediterranean, although we measure INP concentrations at higher temperatures. This is likely related to the fact that GRAINS uses a larger droplet volume (100 µL) than most droplet freezing assays. At UGR, data show median INP concentrations falling in the upper range of the literature data. On the other hand, although INP concentrations at SNS are also falling in the upper region of the data, the variability of the measurements seems to be higher, especially considering the whiskers of the boxplot that reach the lower region of the literature data.

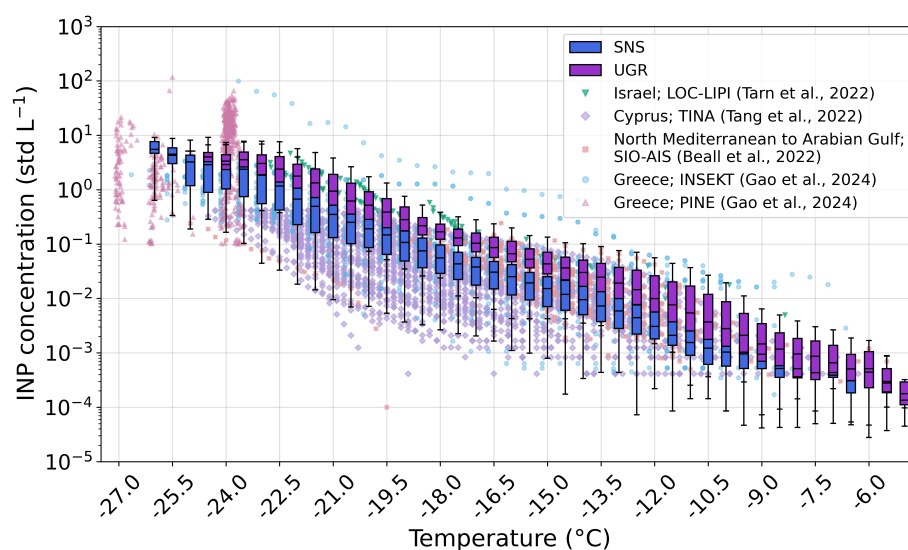


Figure 2. Boxplots of the INP concentration at the UGR and SNS stations for the entire measurement period. Black lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show Q1-1.5IQR and Q3+1.5IQR, respectively. Outliers are not shown for clarity. Literature INP data collected in the Mediterranean region are also shown.



The median INP concentrations range from 1.4×10^{-4} std L⁻¹ at -5 °C to 5.5 std L⁻¹ at -26 °C for SNS, and from 1.8×10^{-4} std L⁻¹ at -5 °C to 4.0 std L⁻¹ at -24.5 °C for UGR. These values fall within the range of atmospheric INP concentrations summarized by Kanji et al. (2017). Nevertheless, differences between measurement sites are expected because
 205 INP concentrations strongly depend on the abundance and composition of aerosol particles. Regarding this, the median INP concentrations observed at the urban UGR site are comparable to those reported for other urban environments, such as Beijing and Mexico City (Chen et al., 2018, 2024; Mendoza-Téllez et al., 2026). In contrast, the SNS measurements are similar to those reported at the high-altitude Mount Helmos station in Greece (Gao et al., 2024) (as seen in Figure 2) and fall within the lower range of observations from the Rocky Mountains in the United States (Zhou et al., 2026). Overall, these comparisons indicate
 210 that the INP concentrations measured at both Mediterranean sites are consistent with observations reported for environments with similar atmospheric characteristics.

The time series of the INP concentration spectra at UGR and SNS is shown in Figure 3, where the red dashed line shows the limit of detection (LOD) of the measurements calculated as proposed by Böhmländer et al. (2025). The INP concentrations range from about 10^{-5} std L⁻¹ to 10^{-3} std L⁻¹ at the highest temperatures, and from 10^0 std L⁻¹ to 10^1 std L⁻¹ at the
 215 lowest ones. Figure 3 shows an increase in the upper limit of INP concentrations measured from January 2025 at both stations, which reflects the implementation of sample dilutions, allowing higher INP concentrations to be quantified. In general, both stations show variations in the INP concentrations depending on the month of the year, although this feature is more evident at SNS. Particularly, at UGR there are lower INP concentrations during the winter season and higher concentrations during spring and summer. At SNS, INP concentrations increase towards the end of summer and decrease during autumn, a pattern
 220 observed in both 2024 and 2025. To gain further insight into the ice nucleation properties at both stations, Figure 4 shows the time series of the INAS densities (n_s), where spectra with filled markers were calculated with the surface area from direct APS measurements, whereas transparent markers correspond to a normalization by the surface calculated with the multiparametric fit explained in Section 3.2. The n_s values range from around 10^3 m⁻² at the highest temperatures to 10^9 - 10^{10} m⁻² at the lowest ones at UGR and SNS, respectively. Overall, at SNS there are systematically higher n_s values than at UGR over most
 225 of the investigated temperature range (see Figure S5). In particular, the n_s range at SNS shows similar value as those reported in previous high-mountain stations (Zhou et al., 2026). From Figure 4, a more pronounced seasonal variability in n_s can be observed compared to the INP concentrations, with higher n_s values during winter and lower values during summer. This behavior is consistent with the enhanced occurrence of mineral dust intrusions during summer in the Western Mediterranean region (Lyamani et al., 2010), which increases the aerosol surface area used to calculate n_s .

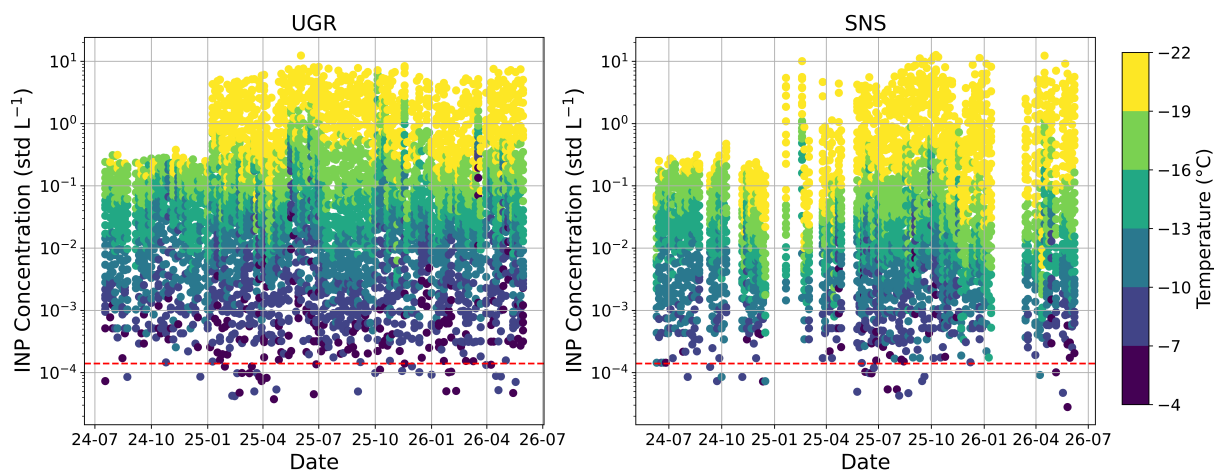


Figure 3. Time series of the INP concentration at the UGR (left) and SNS (right) stations. Red dashed line shows the limit of detection of GRAINS.

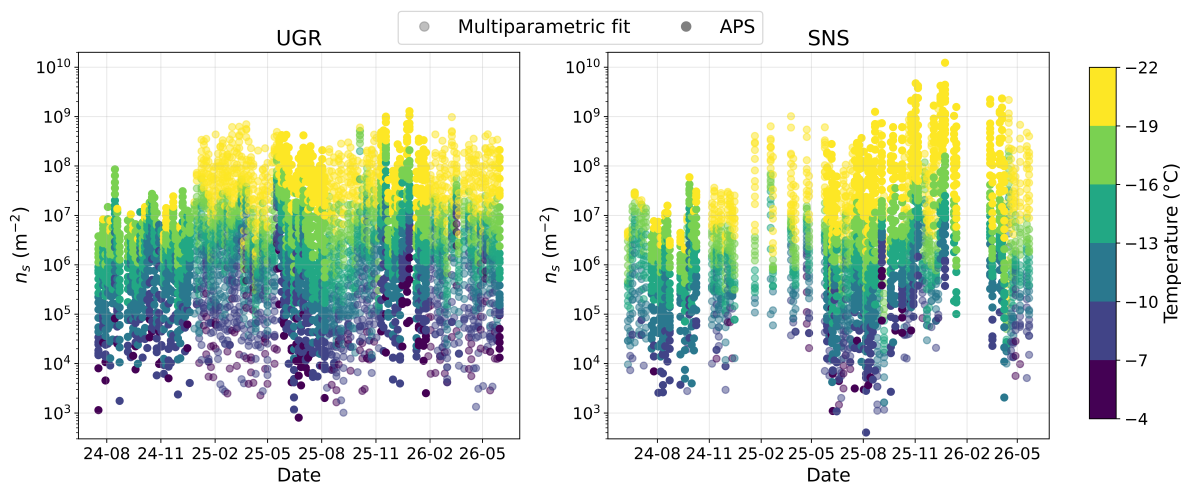


Figure 4. Time series of the INAS density at the UGR (left) and SNS (right) stations. Transparent markers correspond to INAS densities calculated with the surface area determined with the multiparametric fit (3.2), whereas filled markers correspond to INAS densities calculated with the surface area from direct APS measurements.

230 A more detailed seasonal analysis of the ice nucleation properties at both stations is shown in Figure 5 that includes boxplots of the INP concentration in each meteorological season. For this analysis data were binned to 1 °C. Since measurements were planned every 4 days, the maximum number of measurements during each season (3 months of each year) should be approximately 46. Boxes at each temperature that contain less than 30% of this number are not considered for statistics and,



therefore, not shown. On the other hand, boxes that contain a number of measurements between 30 and 50% of this target are
 235 shown with transparency. Normal colored boxes represent a dataset with at least 50% of the target measurements.

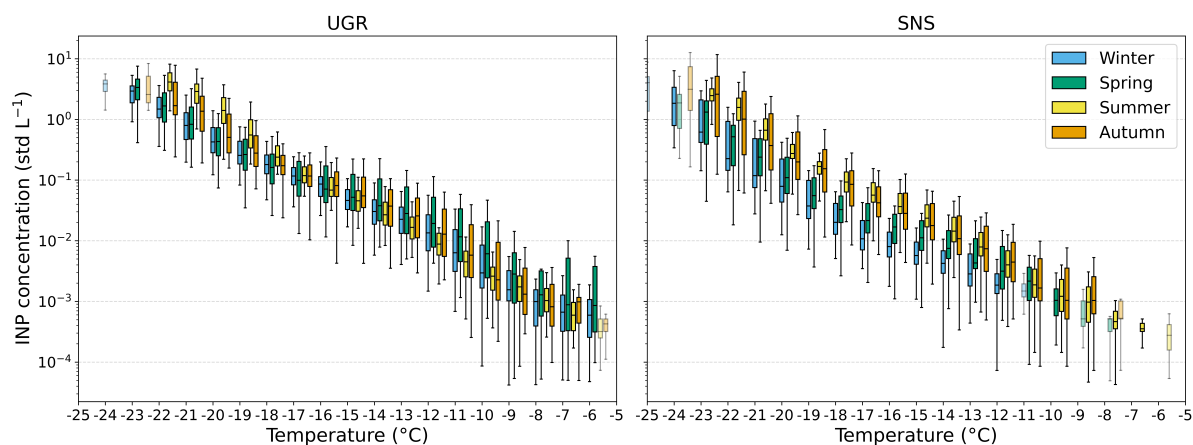


Figure 5. Boxplots of the INP concentration in each meteorological season at UGR and SNS. Black lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show $Q1-1.5IQR$ and $Q3+1.5IQR$, respectively. Outliers are not shown for clarity. Transparent boxes represent datasets with a low number of data points for the statistics.

At UGR, INP concentrations at high temperatures are very similar among seasons, although spring presents higher variability. In fact, when looking at the median INP concentration of each season (better seen in Figure S6) one can observe that during spring the median INP concentration at high temperatures is higher compared to the rest of the seasons. High INP concentrations at warmer temperatures (> -15 °C) are associated with particles of biological origin (Huang et al., 2021) and the spring season coincides with the highest concentration of pollen particles in Granada (Cariñanos et al., 2025). Therefore, the higher INP concentrations shown in Figures 5 and S6 are most likely caused by different pollen or subpollen particles emitted into the atmosphere. Regarding the lower temperature range, summer and autumn present higher INP concentrations. In this temperature range, mineral dust is the aerosol particle that dominates the ice nucleation properties in immersion freezing (DeMott et al., 2015). This is in agreement with more frequent occurrence of Saharan dust intrusions into the Iberian Peninsula (Lyamani et al., 2010). However, at UGR autumn presents lower values compared to summer, which could be related to the presence of other particles at the UGR station that are present in the atmosphere during dust events and affect the ice nucleation properties of the entire aerosol sample. In particular, BC particles are known to be inefficient INPs (Vergara-Temprado et al., 2018) and there is evidence of mixing between dust and BC particles (Scarnato et al., 2015). These interactions of dust with BC and other anthropogenic aerosol particles can change the number of active sites in the particles, so it would affect the ice nucleation properties (Li et al., 2026). The seasonal behavior observed at UGR is consistent with the year-round measurements reported by Tobo et al. (2020) over the Tokyo metropolitan area, one of the few studies describing the seasonal variability of atmospheric INPs at an urban site. They observed relatively small seasonal variations at temperatures below approximately -20
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°C, except during Asian dust events, whereas INP concentrations between approximately -15 and -10 °C were systematically higher during the warm and wet seasons than during the cold season. They attributed this behavior to seasonal changes in particles of biological origin together with episodic contributions from mineral dust, which is in agreement with the seasonal patterns observed at UGR.

At SNS there are higher INP concentrations at warm temperatures during spring, summer and autumn compared to winter. This could be related to the fact that during winter the station is commonly in the free troposphere, so aerosol particle concentrations are very low compared to the other seasons, when there is more influence of particle transport within the PBL. Particularly during spring and summer, Cariñanos et al. (2025) also reported high concentrations of pollen particles at SNS, so high INP concentrations could be explained by the presence of pollen particles in the high-mountain station. The behavior of median INP concentrations at high temperatures during autumn is also likely due to particles from biological origin, although the pollen concentrations shown in Cariñanos et al. (2025) during the autumn season are quite low. A possible hypothesis is that the INPs during this season belong to a different type of biological particles, although with the instrumentation available in this study it is not possible to confirm it. Regarding this, Bras et al. (2024) found higher concentrations at Puy-de-Dôme (France) during the spring and autumn months, with most of the INPs being heat labile and therefore, probably coming from biological origin. As for winter, aerosol particle concentration is usually very low, so the lower values of INP during this season could be due to this fact. Zhou et al. (2026) found lower INP concentrations at the Rocky mountains during the cold seasons (December to March) as compared to the other months, which agrees with our lower INP concentrations during winter (December to February). At low temperatures, summer and autumn show the highest INP concentrations, which is again linked to the more frequent presence of mineral dust in the atmosphere during these seasons (Bazo et al., 2023; Lyamani et al., 2010).

To evaluate the effect of surface on the seasonal variability of ice nucleation properties, we have also analyzed the median n_s in each meteorological season (Figure S7). At UGR, n_s at high temperatures show clear differences among seasons, with higher values during spring and winter compared to summer and autumn. Something similar occurs at SNS, where n_s at high temperatures shows very similar values for spring, winter and autumn and lower values for summer. Since the majority dust events occur during the summer season, these results were expected as a result of a larger surface available for ice nucleation.

4.2 Relationship between ice nucleation properties and optical properties

Despite their critical role in aerosol–cloud interactions, observations of INPs remain sparse and unevenly distributed, limiting our understanding of their abundance, variability, and activation behavior across different atmospheric environments. To address this gap, in this section we explore the relationship between INP concentrations and aerosol optical properties, which are commonly used for aerosol classification (Cazorla et al., 2013; del Águila et al., 2025; Schmeisser et al., 2017). In particular, we applied the Ångström matrix proposed by Cappa et al. (2016) using the optical measurements from the nephelometer and the AE33/36s instruments at both stations. Figures 6 and S8 present the resulting Ångström matrix for UGR and SNS coloured according to the INP concentration and the n_s at -15 °C, respectively. The SAE and AAE correspond to hourly averaged values, whereas both the n_s and the INP concentrations correspond to daily measurements (the same INP concentration is assumed throughout the day).



At UGR, most aerosol categories defined by the Ångström matrix show comparable INP concentrations, with the majority of the observations lying within one order of magnitude (Figure 6). Nevertheless, systematically higher INP concentrations are observed for the Large particle/BC mix and Mixed dust/BC/BrC categories, which are typically associated with mineral dust outbreaks considering the region of study. Conversely, some observations classified as Large particles/Low absorption exhibit the lowest INP concentrations. These cases are likely associated with coarse particles of non-mineral origin, which are generally less efficient ice nucleating particles at these temperatures. Unlike the INP concentrations, the n_s values exhibit a distinct pattern across the Ångström matrix (Figure S8). Lower n_s values are associated with the dust-related categories, which is expected since mineral dust events are dominated by larger particles, resulting in a larger aerosol surface area and therefore lower ice-active site densities. In contrast, the categories dominated by BC exhibit the highest n_s values.

At SNS, the highest INP concentrations are generally associated with the dust-related regions of the matrix, whereas the BC-dominated and low-absorption regions are characterized by lower INP concentrations. This contrasts with the behavior of n_s , where lower values are generally found in the dust-related regions, while the highest values occur for BC and Mixed BC/BrC aerosols. Within the Mixed dust/BC/BrC region, n_s decreases with decreasing SAE and increasing AAE. As shown by Bazo et al. (2025), optically classified Dust cases correspond to mineral dust outbreaks dominated by relatively pure dust particles. Therefore, the trend observed in Figure S8 suggests that the dust events dominated by the purest mineral dust particles are associated with the lowest n_s values. A similar behavior is observed across the remaining aerosol categories, with the highest n_s values corresponding to BC and Mixed BC/BrC aerosols. In contrast, some low-absorption aerosol cases also exhibit relatively low n_s values.

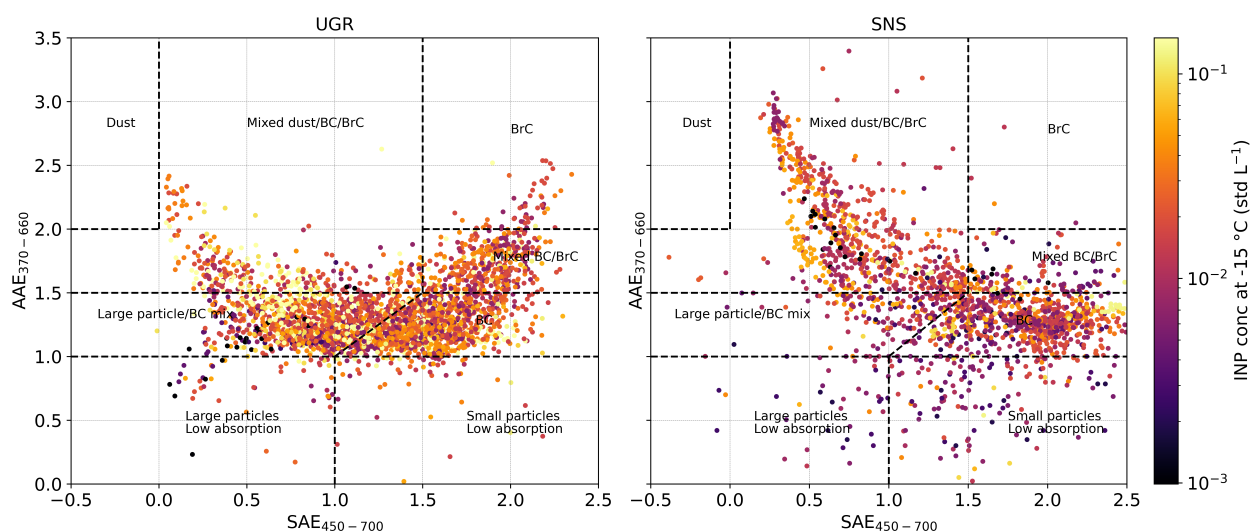


Figure 6. Absorption Ångström exponent (AAE) versus scattering Ångström exponent (SAE) for the entire measurement period. Data points represent hourly averages of the optical properties and are colored by the INP concentration at -15°C . The classification shown was proposed by Cappa et al. (2016).



305 Given the observed relationship between ice nucleation and aerosol optical properties, particularly the SAE and AAE, we
further evaluated the correlation between INP concentrations and several optical parameters to quantitatively assess these
relationships. Table 2 shows the Spearman coefficients between the INP concentrations at -12, -15 and -18 °C and σ_{sp} at 550
nm, σ_{ap} at 370 nm, AAE and SSA, as SAE did not show significant correlations at any station or temperature. We have also
calculated the Spearman coefficients between the INP concentrations and the number particle concentration above 500 nm
310 (N_{500}) as it has shown a strong correlation in many studies (DeMott and Prenni, 2010; DeMott et al., 2015). At UGR, the only
variable that shows correlation with the INP concentrations is the SSA, with negative Spearman coefficients. This behavior
would indicate that there are higher INP concentrations values for less scattering particles, which is linked to the presence
of BC particles in the urban environment. However, the Spearman coefficients are low so the correlation is weak. On the
other hand, at SNS many aerosol properties show significant correlations with the INP concentrations. Positive correlations
315 were found for AAE and SSA, suggesting that aerosol composition influences the observed INP concentrations. The positive
correlation coefficients for AAE and SSA agree with the presence of mineral dust particles. Also, the σ_{sp} and the σ_{ap} show
positive correlations generally above 0.4 (except for the INP concentration at -12 °C). These Spearman coefficients show similar
values as the one obtained for the correlations with N_{500} . Since these three variables are all related to aerosol loading in the
atmosphere, it could indicate that σ_{sp} and σ_{ap} give similar information in terms of relationship with INPs.



Table 2. Spearman correlation coefficients with p -values < 0.05 between the INP concentrations at -12, -15 and -18 °C and several aerosol optical properties. Bold numbers represent p -values below 0.005. N represents the number of measurements involved in the correlation analysis.

Station	Temperature (°C)	Variable	Spearman	N
UGR	-12	SSA (550 nm)	-0.250	153
	-15	SSA (550 nm)	-0.226	148
	-12	σ_{sp} (550 nm)	0.338	97
	-12	σ_{ap} (370 nm)	0.301	90
	-12	SSA (550 nm)	0.238	81
	-15	σ_{sp} (550 nm)	0.463	100
	-15	σ_{ap} (370 nm)	0.479	92
	-15	AAE	0.299	92
SNS	-15	SSA (550 nm)	0.337	85
	-15	N_{500}	0.491	53
	-18	σ_{sp} (550 nm)	0.468	100
	-18	σ_{ap} (370 nm)	0.489	92
	-18	AAE	0.286	92
	-18	SSA (550 nm)	0.334	85
	-18	N_{500}	0.421	53

320 4.2.1 Predicting INP concentrations at UGR and SNS using aerosol properties

INP parameterizations are crucial for representing aerosol-cloud interactions in climate models. To identify aerosol properties suitable for the development of INP parameterizations at UGR and SNS, Random Forest (RF) regression models were applied, with predictor importance assessed using permutation feature importance (PFI). RF is an ensemble machine-learning method that combines multiple decision trees (1000 in this analysis) to model the relationship between predictors and response variables. RF approaches have also been used to develop parameterizations by directly predicting target variables from aerosol properties (Zabala et al., 2026). However, in this study RF was used to quantify the relative importance of aerosol predictors rather than to predict the parameters of the INP parameterization. For each model, 70 % of the data were used to train the RF, while the remaining 30 % were reserved for testing. PFI was calculated on the independent test dataset by randomly permuting each predictor 100 times and quantifying the resulting change in model performance, where larger decreases indicate a greater



330 contribution of the predictor to the model. The analysis was performed at the same temperatures considered in the previous correlation analysis (-12 , -15 and -18 °C). Three RF models were trained by combining either σ_{sp} , σ_{ap} or N_{500} with the intensive optical properties (SAE, AAE and SSA). Since σ_{sp} , σ_{ap} and N_{500} are all proxies for aerosol loading, they were evaluated separately to avoid redundancy among predictors.

For the high-mountain environment, σ_{sp} and σ_{ap} became the dominant predictors, showing feature importance values comparable to those obtained for N_{500} when it was included instead (see Table S3). This suggests that the three variables contain redundant information related to aerosol loading. In contrast, no single variable dominated the RF models at the urban station (Table S4), except for AAE at -15 °C. For the other temperatures, AAE, SSA and SAE showed comparable permutation feature importance values, with some cases where the contribution of aerosol loading proxies (σ_{sp} , N_{500} and σ_{ap}) was smaller. This suggests that, unlike SNS, the variability of INP concentrations at UGR is more closely linked to changes in aerosol composition than to aerosol abundance.

Following the feature importance analysis, we have obtained and evaluated different parameterizations of INP concentrations at SNS and UGR. As in previous parameterizations reported in the literature, we have developed an expression that includes a power-law dependence on the specific aerosol property involved and an exponential dependence on temperatures. The parameterization coefficients were obtained by taking logarithms of the expression and then fitting the equation to a linear regression. For this, we have first used a 70 % of the available dataset to develop the different parameterizations and then we have evaluated them with the remaining 30 % of the dataset, in order to check the performance with an independent subset of observations.

Based on the results of the feature importance for SNS, we developed two different parameterizations based on aerosol loading. The first parameterization predicts the INP concentration spectra as a function of the N_{500} , as typically done in previous studies (DeMott and Prenni, 2010; DeMott et al., 2015; Gao et al., 2024). However, in the second one the INP concentration spectra is obtained as a function of the σ_{sp} . To our knowledge, this is the first study where scattering coefficients are used to predict the INP concentration. Table 3 shows the formulations of the parameterizations proposed, as well as the evaluation performed with the remaining 30 % of the dataset, which can be visualize also in Figure 7. For both parameterizations, we propose a simpler formulation than in previous parameterizations proposed in the literature, since introducing an interaction term between temperature and aerosol loading (N_{500} or σ_{sp}) only caused a very small improvement in the prediction.

The SNS parameterization based in N_{500} shows an r^2 over 0.9 when predicting the INP concentration spectra, with a 68.2 % of data within a factor 2, 98 % of the data within a factor 5 and 99 % of the data within a factor 10. There are only a few points that fall out of the factor 10 region, which correspond to the higher temperature range, where the INP concentration data has larger uncertainties. Figure 7 shows that overall, the parameterization overestimates the measured INP concentration, with a median relative bias of 35.3 %. In particular, at high temperatures, the parameterization slightly overestimates the INP concentrations, as there are more data points above the 1:1 line. On the other hand, in the lower temperature range the INP concentration data are generally below the 1:1 line, so the parameterization underestimates the data. Still, most of these cases are within factor 2 and 5, so the agreement between the predicted and the measured INP concentrations is very good. Concerning the parameterization based on σ_{sp} , the r^2 resulted from the evaluation is below 0.9, but very close to it, with a value



365 of 0.88. This parameterization predicts 65.2 % of the data within a factor 2 compared to the measured INP concentrations, whereas 93.4 % and 97 % of the data are within factors 5 and 10, respectively. For this parameterization there is also a slight overestimation of the INP concentration for the higher temperature range, whereas for the lower temperatures there is again an underestimation of the data. Overall, the median relative bias is -1.8 %, which indicates a negligible underestimation compared to the N_{500} parameterization. As in the N_{500} parameterization, the majority of the data is within a factor 5, so there is good agreement between the predicted and the measured INP concentrations. Bras et al. (2024) and Gao et al. (2024) also developed parameterizations for predicting INP concentrations at high-mountain stations. While Bras et al. (2024) developed one parameterization based on particle number concentration, Gao et al. (2024) developed several considering different microphysical properties. The ability of these parameterizations to predict INP concentrations was evaluated using the fraction of observations falling within different prediction factors, providing a comparable metric to the one used in this study. The parameterization proposed by Bras et al. (2024) predicted 57.3 %, 87.7 % and 98.0 % of the measured INP concentrations within factors of 2, 5 and 10, respectively, while the parameterizations proposed by Gao et al. (2024) generally predicted between 55 and 70 % of the observations within a factor of 3 and around 90 % within a factor of 10. These results are consistent with the predictive performance of the parameterizations developed in this study.

380 The two parameterizations developed for SNS are similar in performance, confirming that aerosol loading is the main driver of INP variability at the high-mountain station. Even though the N_{500} parameterization performs slightly better in terms of r^2 , the one based on σ_{sp} shows a lower median relative bias and could also be applied to larger datasets since it is a more available variable, making it a more suitable predictor in order to develop a general parameterization at SNS. This opens new possibilities for obtaining the INP concentration at stations with similar conditions as SNS, since optical measurements are routinely measured in many atmospheric stations.



Table 3. Parameterizations proposed for SNS and UGR.

Station	Aerosol property	Formulation	Evaluation
SNS	N_{500}	$\text{INP} = 10^{-5.091} N_{500}^{0.131} 10^{-0.225 T}$	$r^2 = 0.92$
			Factor 2 = 68.2 %
			Factor 5 = 98.0 %
			Factor 10 = 99.2 %
	$\sigma_{\text{sp}} (550 \text{ nm})$	$\text{INP} = 10^{-5.553} \sigma_{\text{sp}}^{0.362} 10^{-0.221 T}$	$r^2 = 0.88$
			Factor 2 = 65.2 %
			Factor 5 = 93.4 %
			Factor 10 = 97.0 %
UGR	SSA (550 nm)	$\text{INP} = 10^{-4.314} \text{SSA}^{-0.307} 10^{-0.216 T}$	$r^2 = 0.85$
			Factor 2 = 52.5 %
			Factor 5 = 88.3 %
			Factor 10 = 97.2 %

385 As seen in Table 3, for UGR we have only developed one parameterization based on SSA, which is linked to aerosol composition. Despite the results for the feature importance regarding aerosol loading, we tried to find two parameterizations involving N_{500} and σ_{sp} , respectively. However, the coefficients found in each equation were not statistically significant and practically zero. This suggests that at the urban environment, INP concentrations are more influenced by aerosol particle type than by aerosol loading. Even though the RF model revealed high feature importances for AAE and SAE, the same happened

390 with the coefficients found in the parameterization. For the case of SSA, we found that the evaluation of the parameterization only improved slightly compared to a parameterization that only accounts for the temperature dependence. Still, as in the proposed parameterization from Table 3 the coefficient of SSA was statistically significant and clearly different from zero, we further explored its potential to become a predictor of INP concentration spectra at UGR. Figure 8 shows the scatter plot of the measured INP concentrations at UGR and the predicted INP concentrations obtained with the SSA parameterization. When

395 evaluating the SSA parameterization, the r^2 obtained is 0.85, with a 52.5 % of the data falling in the region delimited by a factor 2. Even though the percentage of data within factor 2 is lower compared to the results obtained for SNS, the percentage increases when considering factors 5 and 10, with 88.3 % and 97.2 % of the data within these factors, respectively. These results are in agreement with the parameterization evaluation by Chen et al. (2026) in an urban environment, with 82.8 % of the data within a factor 5. Figure 8 shows that generally the data fall around the 1:1 line, although there is some deviation

400 for the lower INP concentration values, with a slight overestimation of the INP concentrations by the parameterization, with a median relative bias of 27.8 %. Still, results show good agreement between predicted and measured INP concentrations.

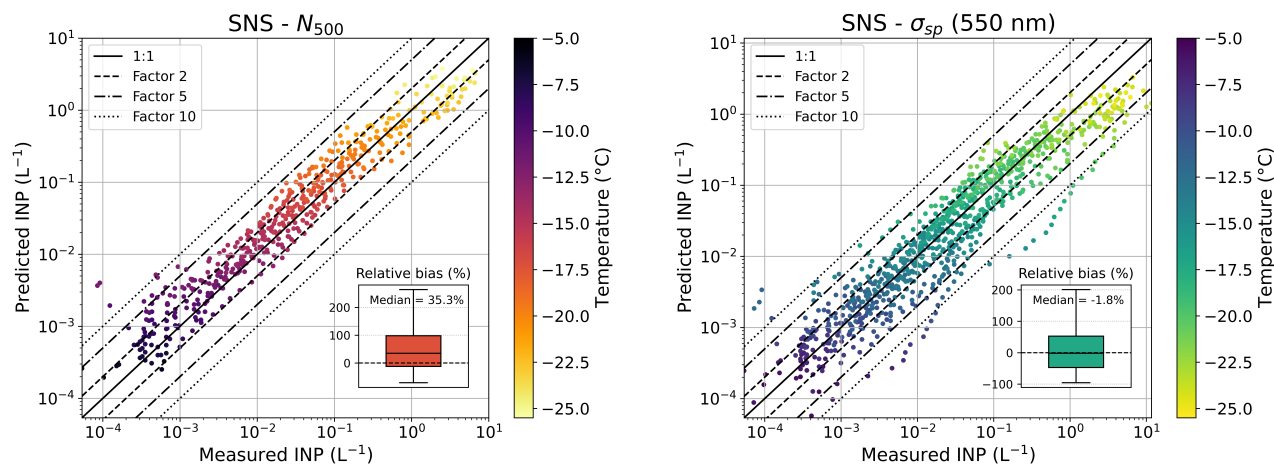


Figure 7. Scatter plots of the measured INP concentration and the INP concentration calculated with the parameterizations based on N_{500} (left) and σ_{sp} (right) at SNS. Solid line represents the 1:1 line, whereas different dashed lines represent agreement factor of 2, 5 and 10. Inset plots show the boxplots of the relative bias, where black lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show Q1-1.5IQR and Q3+1.5IQR, respectively. Outliers are not shown for clarity.

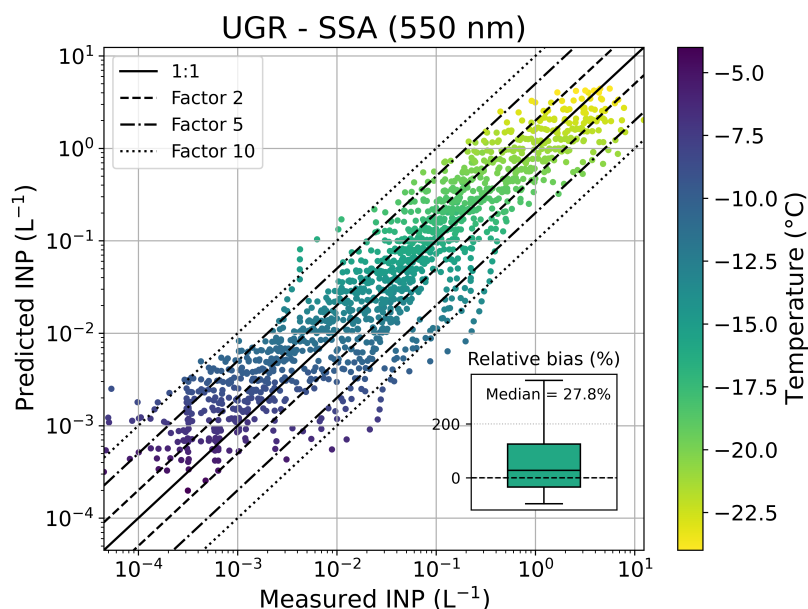


Figure 8. Scatter plots of the measured INP concentration and the INP concentration calculated with the parameterization based on SSA at UGR. Solid line represents the 1:1 line, whereas different dashed lines represent agreement factor of 2, 5 and 10. Inset plots show the boxplots of the relative bias, where black lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show Q1-1.5IQR and Q3+1.5IQR, respectively. Outliers are not shown for clarity.

The results from the evaluation of the INP parameterization based on optical properties suggest that these measurements could be used to estimate INP concentrations in environments similar to those represented by the UGR and SNS stations. Previous studies have demonstrated the potential of optical measurements to predict CCN concentrations at different stations (Shen et al., 2019; Zabala et al., 2026). Since optical measurements are routinely measured in many atmospheric observatories, the approach presented in this study could be further evaluated at other atmospheric stations. This would allow to extend the INP concentration estimates to locations where INP measurements are not available, therefore improving the spatial coverage of INPs around the world.

5 Conclusions

This study presents a two-year characterization (June 2024–June 2026) of ice nucleating particles (INPs) across two contrasting environments in Southern Spain: an urban site (UGR, 37.18 °N, 3.58 °W; 680 m a.s.l.) and a high-mountain station (SNS, 37.10 °N, 3.39 °W; 2610 m a.s.l.). These observations provide a long-term dataset of atmospheric INP concentrations in the Western Mediterranean region, where continuous INP measurements are scarce.



The overview of INP properties at UGR and SNS revealed INP concentrations in the range between 10^{-5} and 10^1 std L^{-1} across a temperature spectrum ranging from $-5^{\circ}C$ to $-26^{\circ}C$, which was consistent with previous INP measurements reported in the Eastern Mediterranean. A clear seasonal variability was observed at both studied sites, with higher INP concentrations in spring at warm temperatures ($> -15^{\circ}C$), related to biogenic emissions. In contrast, summer and autumn seasons showed higher INP concentrations at lower temperatures, associated to the presence of long-range transported Saharan dust in the region. Overall, INP concentrations were systematically higher at UGR than at SNS. On the other hand, at the high-mountain station aerosol particles exhibited a higher ice-nucleation efficiency, as indicated by the density of ice-nucleating active sites (n_s).

The combination of INP measurements with continuous aerosol properties observations provided insight into the drivers of ice nucleation at both stations. At SNS, INP concentrations were mostly related to several proxies of aerosol loading, including the scattering and absorption coefficients and the particle number concentration above 500 nm. However, at UGR, the significant relationship was found for single scattering albedo (SSA), an optical property related to aerosol composition. Permutation feature importance (PFI) analysis supported these results, with aerosol loading variables being particularly relevant for ice nucleation at the high-mountain station, while no aerosol property dominated at the urban station.

Based on these results, different parameterizations based on optical and microphysical properties were developed for both stations. For SNS, a first parameterization was based on N_{500} , whereas another one was based on σ_{sp} . Both parameterizations showed r^2 close to 0.9, with more than 90 % of the data falling within factors of 5 and 10. The parameterization based on σ_{sp} showed a lower median relative bias (-1.8%) than the N_{500} based parameterization (35.3%). At UGR, the SSA based parameterization achieved an r^2 of 0.85, with 88 % and 97% of the observations falling within factors of 5 and 10, respectively.

Overall, these results demonstrate that aerosol optical properties can provide INP parameterizations with a performance comparable to those based on microphysical properties at the urban and the high-mountain station. This suggests that optical properties could be used to estimate INP concentrations in environments similar to the ones investigated in this study, providing an opportunity to extend INP concentration estimations to atmospheric observatories where aerosol optical properties are routinely measured.

Code and data availability. Code and data will be available upon request.

Author contributions. EB conducted GRAINS experiments, performed data curation, formal analysis and investigation and wrote the manuscript. ORG conducted GRAINS experiments and performed data curation and investigation. GT and AC defined the structure of the paper, conceptualized the investigation, supervised the writing of the manuscript and were responsible for the project administration and funding acquisition. All authors contributed to the discussion of the results and provided comments on the paper.

Competing interests. The authors declare no competing interests.



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