



Ice nucleating particle flux density estimated with radon tracer methods during conditions dominated by thermally driven mountain winds

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Abstract. Spatiotemporal variability of tropospheric ice nucleating particle (INP) concentrations, driven by a combination of surface sources and atmospheric transport mechanisms, distinctly modulates their effect on clouds. However, limited understanding of INP emission patterns currently impedes predictions of this variability, particularly for INPs active at little supercooling, e.g. temperatures ≥ -10 °C (INP₋₁₀). Here, we apply two implementations of the radon tracer method (RTM) to quantify the INP₋₁₀ flux density between source regions and a high-altitude site in central Europe during thermally driven planetary boundary layer (PBL) intrusion events. By using observations at a high-altitude site, the chosen implementations of the RTM are applied directly at an altitude where INPs can impact clouds, and at a location where atmospheric conditions regularly change between air masses characteristic of free tropospheric or “baseline” conditions and conditions distinctly influenced by air masses that have experienced recent contact with the terrestrial surface. Median emission flux densities estimated based on our implementation of the traditional RTM using two different radon exhalation maps were approximately 360 INP₋₁₀ m⁻² h⁻¹ and 250 INP₋₁₀ m⁻² h⁻¹, respectively. The median emission flux density modelled in a Bayesian framework was about 720 INP₋₁₀ m⁻² h⁻¹. Initial estimates seem to indicate a limited role of thermally driven transport on fair-weather days in the annual INP₋₁₀ export above the Alps. Relating the calculated flux densities to other kinds of observations, we demonstrate that top-down approaches investigating mean INP flux densities on local to regional scales under particular meteorological conditions can contribute to an improved understanding of INP dynamics at cloud height.

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1 Introduction

Only a minute fraction of aerosol particles can initiate the freezing of supercooled droplets in liquid clouds warmer than -38 °C (Murray et al., 2012); these are referred to as ice nucleating particles (INPs). An important fraction of tropospheric INPs is initially aerosolized from terrestrial surfaces into the PBL (Kanji et al., 2017), which encompasses the turbulently mixed atmosphere extending from the Earth’s surface to heights ranging from 300 m to 3000 m (Stull, 2017). High INP activation temperatures at minimal supercooling (e.g. temperatures ≥ -10 °C; INP₋₁₀) are primarily linked to biological material (Huang et al., 2021; Kanji et al., 2017). The high spatiotemporal variability of such biological INP populations (Kanji et al., 2017) is currently difficult to predict, both, near source regions and at cloud formation levels (Cornwell et al., 2023; Herbert et al.,

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2025). Knowledge on the abundance of INPs with high activation temperatures and their relative fraction of total INPs, is, however, important to understand cloud development and properties (Crawford et al., 2012; Hawker et al., 2021; Kanitz et al., 2011).

During daylight hours under clear-sky conditions, the exchange between the PBL and the overlaying free troposphere (FT) is thermodynamically impaired by the synoptic inversion (Stull, 2017; Williams et al., 2011). Vertical export of PBL air and associated aerosols over flat terrain can occur by mixing and entrainment processes on different spatial scales such as dry and moist convection (e.g. active localized cumulus or large-scale convective thunderstorms (Cotton et al., 1995; Williams et al., 2011)) or frontal uplift (Jin et al., 2022). In each of these cases, surface emissions are integrated from source regions of varying sizes and transported to the FT. Complex terrain fosters additional thermally and dynamically driven uplift, summarized as topographic venting (Henne et al., 2004; Jackson et al., 2013; Zardi and Whiteman, 2013; see Figure 13 in Henne et al. (2004) for a summary of vertical mixing processes above heterogeneous topography). Topographic venting is frequently associated with enhanced PBL injections comprising surface-derived aerosols and trace gases into the FT above mountains (Collaud Coen et al., 2011; Henne et al., 2004; Lugauer et al., 1998; Poltera et al., 2017; Richner and Hächler, 2013). Such periods of PBL influence at the Jungfraujoch Observatory, a high-altitude site in the Swiss Alps, can be identified by the total number of particles ≥ 90 nm, the ratio of NO_y to CO, and increases in absolute humidity or radon-222 activity concentration (Henne et al., 2005; Herrmann et al., 2015; Zellweger et al., 2003).

Radon-222 (henceforth radon) is an unreactive gas generated by the radioactive decay of radium-226 as part of the uranium-238 decay sequence (Porstendörfer, 1994) and a convenient atmospheric tracer of recent (< 2 -3 weeks) terrestrial influence. Compared with most trace gases, radon is emitted continuously and relatively homogeneously from unsaturated soils to the near-surface atmosphere (Nazaroff, 1992), and is readily mixed within the PBL. By contrast, the emission of radon from open water bodies such as oceans is almost negligible (e.g. Zahorowski et al., 2013 and references therein). The combination of radon's short half-life of 3.82 days (Porstendörfer, 1994), and limited vertical exchange typically imposed by the synoptic inversion, leads to significantly lower radon concentrations in the FT than in the PBL over land (Chambers et al., 2013, 2016; Williams et al., 2011).

Air sampled at JFJ is representative of the FT around 40% of the year (Baltensperger et al., 1998; Conen et al., 2022; Herrmann et al., 2015). Periods influenced by PBL intrusions cover about twice as much time in summer as in winter (Herrmann et al., 2015). In winter, PBL intrusions often occur by advection of air mixed vertically upwind of the station, for example during frontal passages, or when synoptic winds are dynamically forced upward by the topography, for example during föhn events (Zellweger et al., 2003). The higher prevalence of PBL intrusions in summer is primarily related to thermally driven uplift, particularly the development of thermally driven mountain winds (Griffiths et al., 2014; Henne et al., 2004; Poltera et al., 2017). Thermally driven mountain winds can develop during anticyclonic conditions characterized by fair weather, clear skies,



strong solar irradiation at the Earth's surface and weak synoptic forcing or large-scale winds. Thermally driven mountain winds can occur along single slopes (anabatic or upslope winds), valleys (up-valley winds), and at larger scales associated with entire mountain massifs (plain-to-mountain winds). Induced by temperature and associated pressure gradients, PBL air is transported upslope toward mountain crests and can be injected into the FT (Henne et al., 2004, 2005; Ketterer et al., 2014; Poltera et al., 2017; Whiteman, 2000; Zardi and Whiteman, 2013). At JFJ, PBL intrusions by anabatic winds are associated with sine-like radon cycles featuring a distinct increase in concentrations in the morning (Griffiths et al., 2014; Lugauer et al., 2000) and occur on about 40% of days between April and September (Griffiths et al., 2014).

Topographic venting at a variety of high-altitude sites and times of the year has been found to coincide with an enhanced number of tropospheric INPs originating from the PBL (Bras et al., 2024; Brunner et al., 2022; Gao et al., 2025; Lacher et al., 2018; Wieder et al., 2022). In this study, we build on these insights by quantifying INP₋₁₀ emission flux densities from the JFJ airshed during periods when the site is influenced by topographic venting. Flux densities are calculated based on two new implementations of the radon tracer method (RTM), originally proposed by Levin (1987). The RTM is a top-down approach that has been implemented in several different ways to estimate greenhouse gas fluxes in a variety of settings and spatial scales (e.g. Biraud et al., 2000; Conen et al., 2002; Gaudry et al., 1990; Grossi et al., 2018; Levin, 1987; Schmidt et al., 1996; Vogel et al., 2012; Wada et al., 2013). Contingent on a range of underlying assumptions, flux densities are inferred by scaling radon flux estimates from a designated fetch region by the ratio of parallel, co-located atmospheric measurements of a species of interest and radon. One implementation of the RTM is the “plume” based approach. It is employed at remote sites where extended periods of recent terrestrial influence characterized by elevated atmospheric radon concentrations - PBL plumes - are interspersed by extended periods of “baseline” or background atmospheric conditions. In such cases, the RTM has successfully been applied to estimate flux densities from terrestrial source regions of varying sizes causing the observed PBL anomalies (Biraud et al., 2000; Gaudry et al., 1990; Gros et al., 1999; Schmidt et al., 1996, 2001; Wada et al., 2013; Wilson et al., 1997). The frequent thermally driven PBL injections at JFJ on fair-weather summer days create similar plume-like events on diurnal timescales (Griffiths et al., 2014). Our implementations of the RTM (Sect. 2.4) during such events offer the possibility to estimate INP₋₁₀ flux densities between an extended, but confined (Lugauer and Winkler, 2005; Weissmann et al., 2005), vegetated source area in the mid-latitudes and an altitude in the atmosphere where INPs can influence clouds during periods of relatively uniform weather conditions that have previously been found to promote aerosol injections above the site (Lugauer et al., 1998; Zellweger et al., 2003).



2 Methods

2.1 Site description

The high-altitude site Jungfraujoch (JFJ) is located on the northern ridge of the Swiss Alps at around 3580 m a. s. l. (Sphinx Observatory, 7.9850° E, 46.5475° N). To the northwest, the terrain slopes steeply towards the mountain pass Kleine Scheidegg (2061 m a. s. l.) and the Swiss Plateau (ca. 400 m to 700 m a. s. l.). The immediate surroundings of JFJ including the area between the site and Kleine Scheidegg are composed of bare rock, covered by the Aletsch glacier to the South-East and, depending on the season, by varying extents of snow. During the sampling period (July-August 2022), the site and parts of the surrounding mountain peaks were covered in snow while the region around and below Kleine Scheidegg was snow-free. Within a radius of 100 km, the land cover around JFJ consists mainly of forest and semi natural areas (69%), with forests, open spaces with little or no vegetation and scrub or herbaceous vegetation associations accounting for 31%, 20% and 18% respectively (Fig. 1). Agricultural areas, artificial surfaces and water bodies cover 22%, 6% and 4% of the area (fractions inferred from the European Union's Copernicus Land Monitoring Service information, 2019).

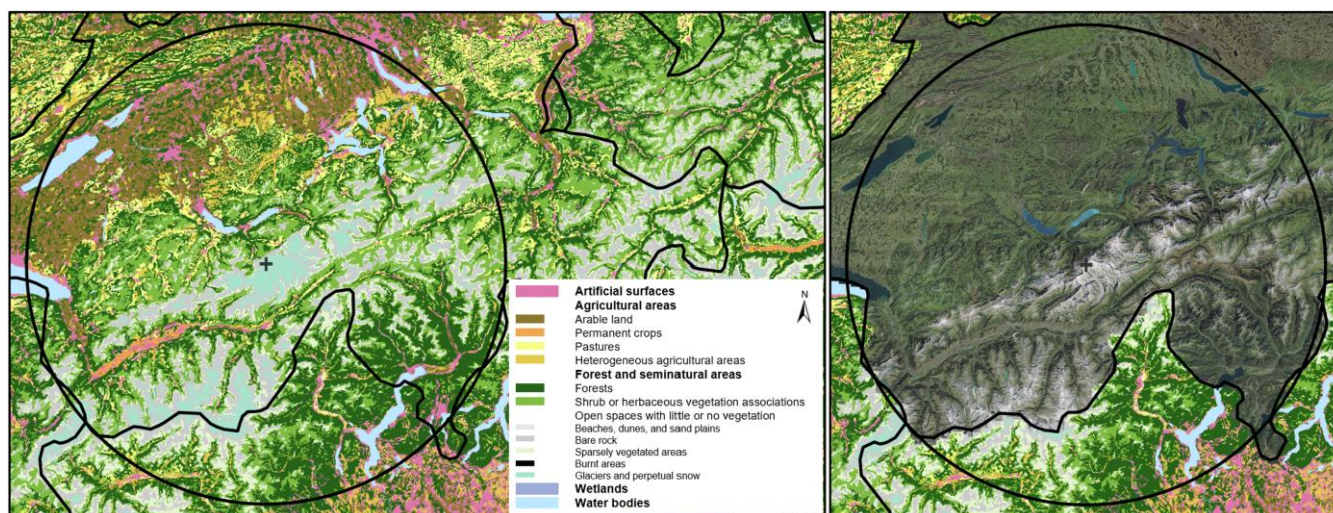


Figure 1: Land cover within a radius of 100 km (black circle) around the high-altitude Observatory Jungfraujoch (black cross, left) superimposed by an Orthophoto of Switzerland (2025, right). Data source land cover: European Union's Copernicus Land Monitoring Service information, <https://doi.org/10.2909/960998c1-1870-4e82-8051-6485205ebbac>. Data source Orthophoto: Swiss Federal Office of Topography (swisstopo).

During the sampling period, winds from northwesterly directions prevailed at JFJ.



115 2.2 Radon and aerosol sampling

Monitoring of 30-minute atmospheric radon concentrations at JFJ commenced in 2009 and is ongoing. Radon measurements used in this study were made with a 700 L dual-flow-loop two-filter radon detector (Whittlestone and Zahorowski, 1998) using a method described by Chambers et al. (2016). Briefly, air is sampled at ~42 LPM and passed through a 400 L delay chamber to enable unwanted radon isotopes (radon-219 and radon-220) to decay. The air is then filtered to remove all ambient radon progeny before passing into the detector's main measurement volume where a consistent proportion of the sampled radon-222 decays and the resulting progeny are filtered for gross α -counting by a zinc sulphide-photomultiplier tube assembly. The detector has a response time of 45 minutes and a lower limit of detection around 0.04 Bq m^{-3} . Automatic calibrations are performed monthly by injecting a known amount of radon from a 21.09 kBq radium-226 source ($\pm 4\%$, Pylon Electronics), and instrumental background checks are made automatically every 3 months. Measurement data is available at Carbon Portal of the Integrated Carbon Observation System (ICOS) (<https://www.icos-cp.eu/data-services>, last access: 16 October 2026).

Aerosol particle concentrations were measured using a white-light optical aerosol spectrometer (Fidas® 100, Palas GmbH, Germany) operated within the Swiss National Air Pollution Monitoring Network (NABEL) on the trace-heated, no-size-cut ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure) inlet. The instrument determines the size distribution in the optical equivalent diameter range between $0.3 \text{ }\mu\text{m}$ and $20 \text{ }\mu\text{m}$ using the side scatter principle and reports them as 1-min averages.

2.3 INP sampling and analysis

We collected a total of 132 INP samples over 23 days between July and mid-August 2022. Sampling targeted periods for which fair-weather conditions were predicted. Starting in the late morning, samples were collected at a temporal resolution of 1.5 h with a liquid impinger (Coriolis μ , Bertin Technologies) at a flow rate of 300 L min^{-1} . The impinger was operated over a period of 30 min per sample at five consecutive 5-min intervals (128 samples) or three consecutive 5-min intervals (4 samples). In between 5-min intervals, water lost from the instrument during sampling was refilled with ultrapure water (W4502-1L, Sigma-Aldrich) to the initial volume of 15 mL. The initial sampling volume consisted of similar ultrapure water supplemented with 5 g NaCl L^{-1} . The addition of NaCl to pure water reduces osmotic stress on biological cells (Stopelli et al., 2014). Furthermore, ion chromatography (940 Professional IC Vario, Metrohm, Switzerland) showed that the initial NaCl content of the impinger liquid diminished to 70% after a sampling duration of 25 min. This indicates a loss of water not solely due to evaporation but also in the form of droplets, which we assume contained at the moment of loss the same INP concentration as the remaining impinger liquid. Assuming that no INPs were lost at the beginning of the collection process from the newly installed cone and a linear increase in INP loss with advancing sampling duration, corrected INP concentrations were obtained by multiplying uncorrected INP concentrations with $\frac{2}{1+0.7}$.



Immediately after sample acquisition, samples were weighed and their INP content quantified using the automatic freezing detection apparatus LINDA (Stopelli et al., 2014). Per sample, usually a total of 52 tubes (0.5 mL Eppendorf Safe-Lock, Germany) provided with 200 μ L sample volume each were analysed. No freezing above -12°C was observed in field blanks comprising NaCl-containing and NaCl-free ultrapure water, respectively.

2.4 Radon Tracer Methods (RTM)

In the following, we present the PBL intrusion formulation of the RTM for INP flux density estimation at JFJ. Figure 2 depicts diurnal radon observations at JFJ on 02 July 2022, a fair-weather day (Sect. 2.4.5) during the sampling period. As characteristic for PBL injections by thermally driven mountain winds at the site, radon concentrations increase in a sine-like pattern between morning and sunset (Griffiths et al., 2014). Starting at sunrise, incoming solar radiation leads to rising temperatures at the Earth's surface and subsequently, the overlaying atmosphere. Upslope winds $> 1 \text{ m s}^{-1}$ can develop within less than one hour (Farina et al., 2023; Nadeau et al., 2020) due to the resulting horizontal temperature contrast between the air above mountain flanks and less elevated terrain. Throughout the morning, turbulence within a growing convective boundary layer as well as wind speed and the thickness of the upslope wind layer increase. Eventually, the upslope winds can get strong enough to break through the capping inversion at the top of the PBL and inject air into the overlaying FT (Henne et al., 2004; Poltera et al., 2017; Zardi and Whiteman, 2013). The onset of a thermally driven flow arriving at JFJ is visible in increasing radon concentrations measured at the station (Griffiths et al., 2014, Fig. 2). On fair-weather days during the sampling period, this arrival was typically observed several hours after sunrise. Over the next few hours the relative proportion of PBL to FT air at the site increases. As previously noted by Griffiths et al. (2014), by early afternoon observations at the station are increasingly similar to PBL air typical of the lower-lying regions of the JFJ airshed. After sunset, cooling of the Earth's surface and the adjoining air causes the upslope flow to diminish, ceasing further PBL injections. In the following, radon concentrations can remain elevated throughout the night, for example in the presence of a residual layer under calm conditions or decrease as a consequence of mixing or an establishing downslope flow drawing down air with little recent terrestrial influence to the Observatory (Griffiths et al., 2014; Ketterer et al., 2014; Lugauer et al., 2000; Whiteman, 2000).

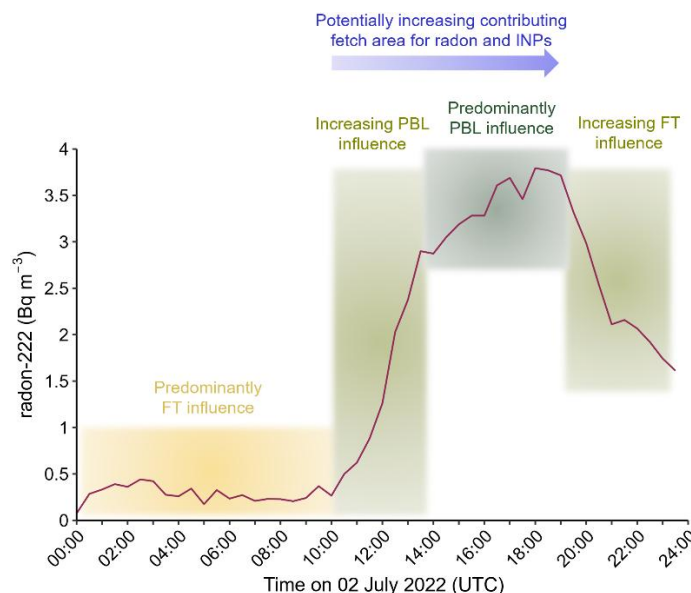


Figure 2: Example of diurnal radon concentrations measured at the high-altitude site Jungfraujoch during a thermally driven PBL intrusion event in summer 2022 (02 July 2022). During the sampling period, sunrise on the Swiss Plateau occurred around 04:00 UTC.

Consequently, thermally driven PBL intrusions are often characterized by a diurnal radon plume-like event, depicting an extended period of recent consistent terrestrial influence and enhanced radon concentrations following a period dominated by FT conditions with limited recent terrestrial influence and low radon concentrations (Fig. 2). In parallel with the observed radon behavior, diurnal aerosol observations at JFJ under conditions associated with PBL intrusion events typically show similar characteristics.

By using radon observations as a source of information about atmospheric transport and mixing, and well-characterized radon emissions, there is enough information to combine INP observations with radon in a model of these plume events to estimate INP emission flux densities. Here, we consider two different models based on different assumptions for interpreting the intrusion events and deriving INP flux densities, which are described below.

2.4.1 Implementation of the traditional RTM for plume-like events

Consistent with the traditional plume RTM application described by Biraud et al. (2000), the mean flux of INP₋₁₀ into the local lower troposphere associated with radon plume events as described in Sect. 2.4 can be estimated according to Eq. (1):

$$F_{\text{INP}_{-10}} = F_{\text{Rn}} \cdot \frac{\Delta \text{INP}_{-10}}{\Delta \text{Rn}} \cdot e^{-\lambda_{\text{Rn}} t} \quad (1)$$

where F_{Rn} describes the mean radon exhalation rate in the source region of the PBL intrusion present at JFJ, ΔRn and ΔINP_{-10} the temporal change of ambient radon and INP₋₁₀ concentrations at the site and λ_{Rn} the radioactive decay constant of radon.



190 At JFJ, ambient radon concentrations $< 0.64 \text{ Bq m}^{-3}$ (local conditions) are indicative of FT or atmospheric background conditions (Conen et al., 2022). During such conditions, filter samples collected across different seasons of the year showed median INP_{-10} concentrations around 2.4 m^{-3} (Conen et al., 2022). This is consistent with the present study where we found a median of $2.9 \text{ INP}_{-10} \text{ m}^{-3}$ when radon concentrations were below 0.64 Bq m^{-3} ($n = 18$). Due to the significant difference between radon and INP_{-10} concentrations in the FT and their respective diurnal peak concentrations during periods affected by PBL intrusions (Fig. 4), ΔRn and ΔINP_{-10} can be approximated by Rn and INP_{-10} . Owing to the short time span between emission and measurement ($< 10 \text{ h}$, Sect. 2.4.4), the radioactive decay of radon during transport can be neglected with an associated error below 5% (Schmidt et al., 2001). The mean INP_{-10} flux into the PBL can then be written as:

$$F_{\text{INP}_{-10}} = F_{\text{Rn}} \cdot \frac{\text{INP}_{-10}}{\text{Rn}} \quad (2)$$

where $\frac{\text{INP}_{-10}}{\text{Rn}}$ corresponds to the slope of a linear regression line fitted to parallel INP_{-10} and radon measurements.

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In other words, mean INP_{-10} flux densities are estimated based on the concept that the ratio of ambient concentration changes of INP_{-10} and radon is proportional to the ratio of their mean flux densities from a mutual source region. Assumptions inherent to the application of the RTM are discussed in detail for the context of the RTM implementations in this study in Sect. 2.4.4.

2.4.2 Box model approach in a Bayesian framework

205 Another option to describe concentration changes of radon and INP_{-10} during an anabatic radon plume event at JFJ as displayed in Figure 2 is with a two-reservoir box model in which radon and INP_{-10} concentrations at the site consist of time-dependent contributions from the FT and PBL air mass (Figure 3). The concentration of radon or INP_{-10} at JFJ at the time t can then be modeled as:

$$c_{\text{JFJ}}(t) = \alpha(t) c_{\text{PBL}}(t) + \{1 - \alpha(t)\} c_{\text{FT}}(t) \quad (3)$$

210 where $\alpha(t)$ [$0 \leq \alpha(t) \leq 1$] denotes the time-dependent fraction of PBL air present at the site, and c_{FT} and c_{PBL} the concentration of the respective substance in the FT background and PBL air mass.

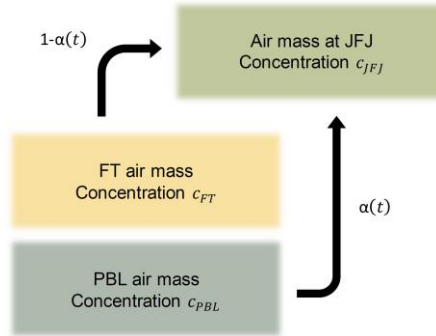


Figure 3: Box model in which atmospheric concentrations of a substance at JFJ are defined by a time dependent contribution from the FT and the PBL air mass.

In a well-mixed PBL, the concentration of both, radon and INP_{-10} , depends on the flux density at the Earth's surface, the mixing height h of the PBL and atmospheric mixing, transport and removal processes. In the absence of horizontal air mass exchange and vertical mixing with the FT, the concentration C_{PBL} of a passive tracer in the PBL air column varies with its initial concentration C_{PBL} at $t = 0$, h , and the mean flux density F since $t = 0$ according to:

$$C_{PBL}(t) = C_{PBL}(t_0) + \frac{F}{h}t \quad (4)$$

The concentration in the FT is set to a constant value, which is equivalent to assuming that changes in FT tracer concentrations occur slowly compared with the timescale on which the model is used (one day). To estimate mean flux densities of INPs in the JFJ catchment during anabatic radon plume events using the framework presented in Eq. (3), we define t_0 as time when the thermally driven flow arrives at JFJ. Based on Eq. (4), Eq. (3) can then be written as:

$$C_{JFJ}(t) = \alpha(t) \left\{ C_{PBL}(t_0) + \frac{F}{h}t \right\} + \{1 - \alpha(t)\} C_{FT}(t) \quad (5)$$

where C_{JFJ} , C_{PBL} and C_{FT} designate atmospheric concentrations of a passive tracer at JFJ, in the PBL air mass and in the FT background air mass, respectively. Based on underlying assumptions discussed in detail in Sect. 2.4.4, the model in Eq. (5) can be used to determine radon and INP_{-10} concentrations at JFJ on fair-weather summer days during anabatic radon plume events. Accounting for the radioactive decay of radon (Porstendörfer, 1994) with the radioactive decay constant λ_{Rn} , the concentrations of radon and INP_{-10} at JFJ can be inferred from:

$$C_{JFJ, \text{INP}_{-10}}(t) = \alpha(t) \left\{ C_{PBL, \text{INP}_{-10}}(t_0) + \frac{F_{\text{INP}_{-10}}}{h}t \right\} + \{1 - \alpha(t)\} C_{FT, \text{INP}_{-10}}(t) \quad (6)$$

$$C_{JFJ, Rn}(t) = \alpha(t) \left\{ C_{PBL, Rn}(t_0) e^{-\lambda_{Rn}t} + \frac{F_{Rn}}{h\lambda_{Rn}} (1 - e^{-\lambda_{Rn}t}) \right\} + \{1 - \alpha(t)\} C_{FT, Rn}(t) \quad (7)$$



with F_{Rn} in Eq. (7) is known (Sect. 2.4.3).

The model in Eq. (6) and Eq. (7) was fitted to observational radon and INP₋₁₀ data measured at JFJ during periods featuring
 235 anabatic radon plume events as defined in Sect. 2.4.5. using the Python library PyMC in a Bayesian framework to model mean
 INP₋₁₀ flux densities during the respective periods. Weakly informative priors were assigned based on previous observations
 and analyses: a PBL height of 1350 m a. g. l. on fair-weather summer days above the Swiss Plateau (Collaud Coen et al.,
 2014), background INP₋₁₀ and radon concentrations of 2 m⁻³ and 0.4 Bq m⁻³ in FT air masses (Conen et al., 2022), background
 INP₋₁₀ and radon concentrations of 11 m⁻³ (Conen et al., 2023) and 1.5 Bq m⁻³ in PBL air masses (Conen et al., 2022), and
 240 radon and INP₋₁₀ flux densities of 80 Bq m⁻² h⁻¹ (Sect. 2.4.3) and 200 m⁻² h⁻¹ (Table 2) in the JFJ airshed.

2.4.3 Mean radon exhalation rates in the source area of PBL intrusions at JFJ

Radon exhalation rates required for the application of the RTM were derived from two process-based monthly European radon
 flux maps available at the ICOS Carbon Portal (Karstens and Levin, 2023). Radon fluxes are modelled based on soil properties
 and soil uranium content combined with either the GLDAS-Noah v2.1 (Karstens and Levin, 2024a) or the ERA5-Land
 245 (Karstens and Levin, 2024b) soil moisture reanalysis dataset. We included the maps for July and August 2022 from both model
 configurations in our analysis. A comparison of radon exhalation rates as calculated by an earlier version of the model with
 other published maps and observational data can be found in Karstens et al. (2015). An evaluation of the updated version of
 the model and the resultant maps used here is available in the corresponding documentation (Karstens and Levin, 2023). It
 suggests that exhalation rates in Central Europe might be up to twice as large as indicated by the maps. For large-scale averages,
 250 radon exhalation rates based on GLDAS-Noah v2.1 might better represent real fluxes than maps based on ERA5-Land
 (Karstens and Levin, 2023). Spatially averaged radon exhalation rates were calculated for all sampling days fulfilling all
 validation criteria for the application of the RTM (RTM days hereafter, Sect. 2.4.5) for source regions determined as described
 in the next section. The results are displayed in Table. 2. Mean radon exhalation rates on RTM days were 90±5 Bq m⁻² h⁻¹
 (mean±SD) and 63±5 Bq m⁻² h⁻¹ (mean±SD) for the maps based on the GLDAS-Noah v2.1 (Karstens and Levin, 2024a) and
 255 the ERA5-Land (Karstens and Levin, 2024b) soil moisture datasets, respectively.

2.4.4 Assumptions made in the implementation of the RTM

- i) Radon and INP₋₁₀ have the same, spatially distributed source area.

The majority of airborne INP₋₁₀ is associated with biological material (Conen et al., 2022; Kanji et al., 2017). Both, emission
 rates of biological INPs and radon exhalation rates from terrestrial surfaces typically exceed those from oceans by orders of
 260 magnitude (Broecker et al., 1967; Huang et al., 2021; Zahorowski et al., 2013). Predominant sources of INP₋₁₀ are vegetation
 (Lindemann et al., 1982; Lindow et al., 1978; Schnell and Vali, 1976) and soils rich in organic matter (Conen et al., 2011;



Fröhlich-Nowoisky et al., 2015; O’Sullivan et al., 2014) where INPs appear to be widespread. As described in Sect. 1, the main sources of atmospheric radon are soil surfaces (Nazaroff, 1992). Combined, this arguably renders (vegetated) soil surfaces the main and mutual source area of radon and INP_{-10} in the JFJ airshed.

265 During PBL intrusion events predominantly induced by thermally driven mountain winds, the source region of the intrusions at JFJ potentially increases from the onset of the event until the flow subsides around sunset (Fig. 2). A systematic difference between radon and INP_{-10} emissions from near and more distant sources within the JFJ airshed could therefore affect the estimated INP_{-10} flux densities. The distinct time lag observed between sunrise and the arrival of the thermally driven flow at JFJ indicates that the first PBL injections already integrate emissions from a wider source area containing air of a convective
 270 boundary layer which had developed over several hours, while the contribution from more distant sources within the catchment might still be minor.

The potential maximal spatial extent of the PBL source area during thermally driven plume events at JFJ depends on horizontal air mass movement in the airshed. For the individual RTM days, we approximated this potential source area extent by drawing a circle centered around the Sphinx Observatory. The circle radii were calculated by multiplying mean wind speed measured
 275 at 51 meteorological stations across Switzerland operated by the Swiss Federal Office of Meteorology and Climatology (MeteoSwiss, Fig. A1) between 00:00 UTC and 19:00 UTC with the length of the time period between the first and last INP sample collected on the individual days. The period between 08:00 UTC and 19:00 UTC corresponds to the first and last INP sample collected after the onset of a distinct increase in radon concentrations (Sect. 2.4.5) considering all RTM days collectively. The maximum time span between the first and last INP sample collected during this period on RTM days was 9
 280 hours. The calculated radii ranged from 43 km to 97 km (Table 2) and are in good agreement with earlier findings revealing that the area from which PBL air is exported by thermally induced topographic venting can extend as far as 100 km into the lowlands (Lugauer and Winkler, 2005; Weissmann et al., 2005). Hence, increases in radon and INP_{-10} concentrations on RTM days likely originated predominantly from the PBL within 100 km of JFJ.

In the absence of strong winds aerosolizing snow from the ground, the bare rock, ice and snow surfaces surrounding JFJ are
 285 neither significant sources of INP_{-10} nor radon. For both substances, sources emerge roughly 1500 m below the site at the transition from bare rock to vegetated terrain. Modelled radon exhalation rates do not vary systematically with distance from JFJ (Fig. A2). Overall, different land cover types within 100 km from JFJ are relatively homogeneously distributed and vary on short spatial scales. The transition between the northern flank of the Alps and the Swiss Plateau (Beyeler et al., 2021) is marked by an increase in the fraction of arable land and a decreasing presence of forest and seminatural areas (Fig. 1, left).
 290 Irrespective of the land cover type, soil surfaces, including agricultural areas, in the JFJ catchment below about 2000 m are usually fully covered by vegetation during summer (Fig. 1, right). Investigations of biological INPs on leaf surfaces of various plant species revealed no systematic difference in their abundance between crops and broadleaved trees (Lindow et al., 1978) or between different broadleaved tree species in a forest in northwestern Switzerland (Einbock and Conen, 2024). Nevertheless, it is possible that INP_{-10} emissions vary with distance from the JFJ which has to be considered when interpreting the flux
 295 density estimates.



ii) Emission rates of radon and INP_{-10} covary and are constant over time.

300 For implementing the RTM in the context of this study, assumption ii) is primarily relevant on diurnal timescales. Temporal variations in radon exhalation rates are mainly driven by changes in soil moisture content (Karstens et al., 2015; Maier et al., 2025; Stranden et al., 1984) and negligible on timescales < 1 day in the absence of significant rainfall. This is typically the case for anticyclonic conditions giving rise to thermally-induced flow (Chambers et al., 2025). Knowledge on the temporal variability in emission rates of biological INPs is currently still limited (Burrows et al., 2022). However, studies have shown
 305 that the emission of biological INPs from certain ecosystems can follow diurnal (Lindemann and Upper, 1985) to seasonal (Creamean et al., 2025; Schneider et al., 2021; Tobo et al., 2020) cycles imposed by variations in environmental parameters and particle inventories. Distinct enhancements in the emission of biological INPs on short timescales can be induced by agricultural activities (Suski et al., 2018) and sometimes coincide with changes in meteorological parameters (Testa et al., 2021; Wright et al., 2014). Particularly rainfall on vegetated surfaces seems to significantly trigger the release of biological
 310 INPs (e.g. Conen et al., 2017; Huffman et al., 2013; Mignani et al., 2025; Testa et al., 2021). Consequently, INP_{-10} source activities cannot be described as constant over time in general terms. Yet, given the prevailing fair-weather conditions during sampling (Sect. 3), the finite sampling periods from late morning to afternoon, and the limited air mass source area integrating emissions across the wider JFJ airshed we expect that possible variability in INP_{-10} emissions is unlikely to undermine the general applicability of the RTM for estimating INP_{-10} flux densities in the present context.

315

iii) Radon and INP_{-10} are mixed, transported and removed from the atmosphere with the same efficiency between their sources and the sampling site.

As a noble gas, radon is efficiently mixed and transported within the atmosphere. Its primary sink is radioactive decay (Porstendörfer, 1994). The dispersal of aerosol particles within the turbulent PBL is influenced by particle aerodynamic
 320 properties and the properties of the present mixing and transport processes (Morris et al., 2013). Particles with aerodynamic diameters around $1 \mu\text{m}$ are well mixed within the PBL, remain typically airborne for multiple days and are mostly removed by wet deposition (Jaenicke, 1980; Wéry et al., 2017). With increasing particle size, the transport efficiency decreases due to higher sedimentation velocities. Depending on wind speed and updraft velocities, particles $> 10 \mu\text{m}$ can be dispersed over distances well exceeding 100 km (Wéry et al., 2017). Our approach quantifies the number of INP_{-10} emitted within the air mass
 325 source region that is successfully transported to JFJ, an altitude in the atmosphere where mixed-phase clouds at -10°C can form. The emission flux of INP_{-10} into the turbulent PBL at their sources might be larger due to INP loss during atmospheric transport.



Overall, assumptions i) – iii) are likely to be met to a sufficient extent for the implementation of the RTM during PBL intrusions at JFJ on the limited temporal and spatial scales investigated in this study.

2.4.5 Validation criteria

Targeted plume events were increases in radon concentrations that resulted primarily from thermally driven PBL injections. As described above, these can be identified based on characteristic sine-like radon cycles at JFJ featuring a distinct increase in radon concentration in the morning on fair-weather days (Griffiths et al., 2014; Lugauer et al., 2000). We approximated the onset time of a distinct and potentially anabatically induced radon increase using a moving-window linear regression approach. The slope of a linear regression fitted to four consecutive, half-hourly radon measurements was calculated within a rolling window between 06:00 UTC and 20:00 UTC. For each day, the maximum slope was identified and the onset of a distinct radon increase defined as the starting time of the first window in which the slope reached $\geq 37\%$ of the daily maximum slope. For estimating flux densities, radon and INP measurements from this time onward were considered.

For the application of the RTM, we considered days with mean daily total sunshine durations > 9 hours and mean daily global radiation $> 220 \text{ W m}^{-2}$ as derived from 51 meteorological stations across Switzerland (adopted from Henne et al. (2004, 2005) and Lugauer and Winkler (2005); validation criteria (a)). These 51 meteorological stations are the same stations we obtained the wind speed data from (Fig. A1, Sect. 2.4.4). All meteorological data is available at <https://www.meteoswiss.admin.ch/services-and-publications/service/open-data.html>, last access 16 October 2026).

The application of the RTM is only reasonable for events with a good correlation between the concentration of radon and the substance of interest. In accordance with previous studies, we did not estimate INP_{-10} flux densities for events with a correlation coefficient $r \leq 0.5$ between INP_{-10} and radon and a p-value ≥ 0.2 ((Schmidt et al., 1996; Wada et al., 2013), validation criteria (b)). To ensure that increases in radon concentrations at JFJ resulted from PBL intrusions, we further excluded days with a correlation coefficient $r \leq 0.85$ between absolute air humidity (Henne et al., 2005) and radon at the Observatory (validation criteria (c)).

3 Results and Discussion

Seven days across the sampling period fulfilled all validation criteria (Sect. 2.4.5) for applying the RTM to estimate INP_{-10} flux densities (henceforth RTM days, Table 1). The other 16 days failed mostly because of too weak a correlation between INPs and radon. In Sect. 3.1, we focus exclusively on the seven RTM days.



Table 1: Number of INP samples per day after the onset of the potentially thermally induced increase in radon concentrations (n), slope of a linear regression fitted to parallel INP₋₁₀ and radon measurements (m) and results of Pearson correlation analyses (correlation coefficient r and p-value) for INP₋₁₀ and radon concentration as well as absolute air humidity and radon concentration at the JFJ for all sampling days excluding one day with < 3 measurements. Top: Seven sampling days fulfilling all validation criteria (Sect. 2.4.5) for the application of the RTM.

Date	n	INP ₋₁₀ – radon			H ₂ O – radon		Failed criteria
		m	r	p	r	p	
2022-07-02	5	1.05	0.86	0.06	0.93	0.02	
2022-07-09	5	13.69	0.85	0.07	1.00	0.0003	
2022-07-14	6	5.41	0.81	0.05	0.98	0.001	
2022-07-18	6	3.92	0.65	0.17	0.97	0.001	
2022-07-19	6	1.97	0.77	0.07	0.97	0.001	
2022-08-03	7	4.43	0.76	0.05	0.87	0.01	
2022-08-13	7	2.24	0.82	0.02	0.96	0.001	
2022-07-01	5	1.45	0.83	0.08	1.00	0.0001	a
2022-07-08	4	-53.69	-0.24	0.76	0.60	0.40	b, c
2022-07-10	4	5.10	0.56	0.44	0.99	0.01	b
2022-07-15	6	1.61	0.39	0.45	0.73	0.10	b, c
2022-07-16	5	0.63	0.39	0.51	0.98	0.004	b
2022-07-17	5	-13.97	-0.76	0.13	0.99	0.002	b
2022-07-26	7	4.17	0.47	0.29	0.38	0.41	a, b, c
2022-07-27	7	-7.13	-0.66	0.11	-0.32	0.48	a, b, c
2022-07-28	4	-10.14	-0.79	0.21	0.81	0.19	b, c
2022-08-04	8	-0.43	-0.07	0.88	0.98	0.00001	b
2022-08-05	4	-8.15	-0.42	0.58	-0.19	0.81	a, b, c
2022-08-06	7	3.21	0.56	0.19	0.95	0.001	a
2022-08-07	3	-4.18	-0.89	0.30	-0.15	0.91	a, b, c
2022-08-12	3	6.33	0.35	0.77	0.52	0.65	b, c
2022-08-14	5	0.60	0.40	0.51	0.72	0.17	a, b, c

Validation criteria:

- (a) Mean daily total sunshine duration > 9 hours or mean daily global radiation > 220 W m⁻² as calculated from 51 meteorological stations across Switzerland (adopted from Henne et al. (2005) and Lugauer and Winkler (2005))
- (b) Correlation coefficient r between INP₋₁₀ and radon > 0.5 and p < 0.2 (Schmidt et al., 1996; Wada et al., 2013)
- (c) Correlation coefficient r between absolute humidity and radon at JFJ > 0.85

3.1 RTM days

On RTM days, Switzerland experienced high mean daily total sunshine durations (≥ 12 hours) and mean daily global radiation (≥ 305 W m⁻²) as observed at 51 meteorological stations across the country (Sect. 2.4.4, Fig. A1). Simultaneously, daytime mean wind speed between 08:00 UTC and 19:00 UTC was ≤ 3 m s⁻¹. At JFJ, radon concentrations and absolute humidity



increased distinctly between morning and afternoon (Fig. 4). Radon concentrations usually peaked in the late afternoon or evening. In some cases, concentrations remained elevated or increased further during the night. These observations indicate that thermally driven mountain winds were indeed a major driver of PBL intrusions at JFJ on RTM days (De Wekker and
375 Kossmann, 2015; Griffiths et al., 2014; Henne et al., 2004, 2005; Lugauer et al., 2000; Whiteman, 2000; Zardi and Whiteman, 2013). Meteorological conditions across Switzerland together with the diurnal profiles of radon and absolute humidity suggest that the diurnal increases in INP_{-10} concentrations observed here were triggered to a significant part by thermally induced PBL air injections entraining INPs from sources predominantly within the radii indicated in Table 2. Diurnal increases in INP_{-10} concentration of a comparable magnitude as observed here have also been observed for advection-induced orographic lifting
380 at Weissfluhjoch (2693 m a. s. l., Swiss Alps) in late winter (Wieder et al., 2022).

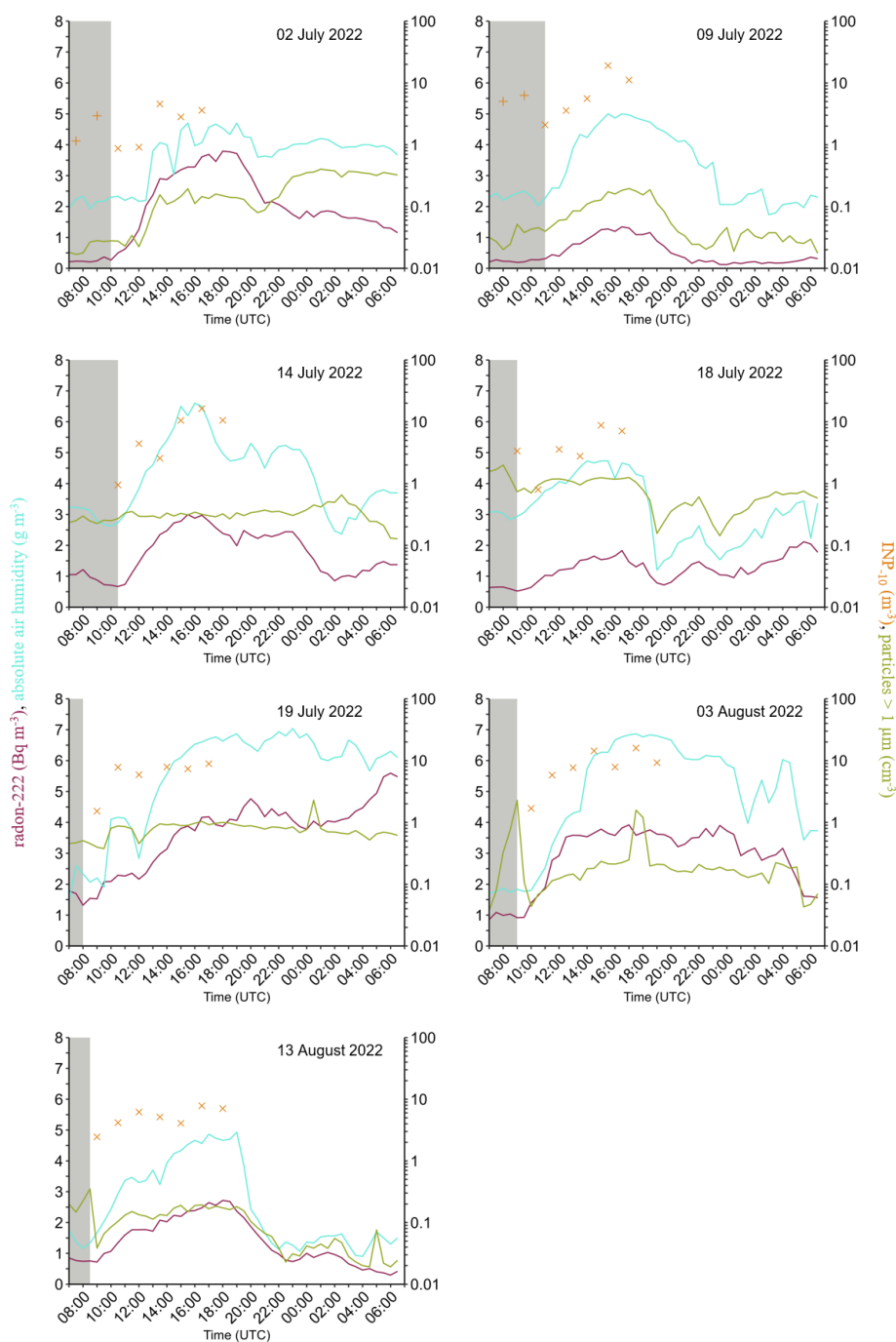


Figure 4: Temporal development of the radon concentration (purple), absolute humidity (blue) and number of particles $> 1 \mu\text{m}$ (green) on RTM days at JFJ. Orange crosses and saltire display INP_{-10} concentrations before and after the onset of a thermally driven radon increase, respectively. Grey shaded area marks the time before the thermally driven increase in radon concentrations (Sect. 2.4.5). Days that only failed due to a too weak correlation between radon and INP_{-10} are shown in Fig. A3.



3.1.1 INP flux density estimates

The slope of a linear regression line fitted to the parallel radon and INP₋₁₀ measurements on each of the seven RTM days ranged from $1.05 \frac{\text{INP}_{-10}}{\text{Bq}}$ to $13.69 \frac{\text{INP}_{-10}}{\text{Bq}}$ (Table 1 top, Fig. 5 (a)) with a median of $3.92 \frac{\text{INP}_{-10}}{\text{Bq}}$. When all data points were combined (n = 42) and binned by radon concentration (11 bins, width = 0.352 Bq m^{-3} , first bin starting at 0.308 Bq m^{-3} , Fig. 5 (b)) or by hour-of-day (11 bins, Fig. 5 (c)) the slopes fitted to the bin median values were smaller, $1.83 \frac{\text{INP}_{-10}}{\text{Bq}}$ and $2.79 \frac{\text{INP}_{-10}}{\text{Bq}}$, respectively.

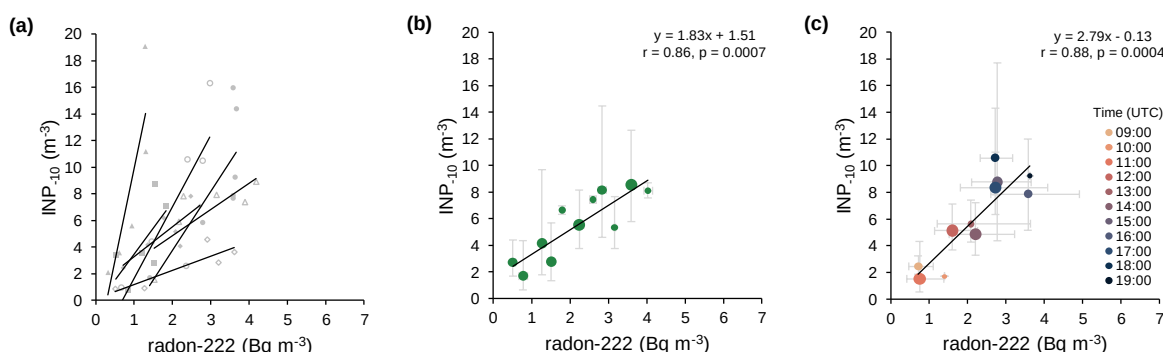


Figure 5: (a) Linear regression lines (black) between parallel radon and INP₋₁₀ measurements for the seven sampling days (symbols) fulfilling all validation criteria for the application of the RTM (RTM days; Sect. 2.4.5). Linear regression fitted to median radon and INP₋₁₀ concentrations on these seven days binned by (b) radon concentration (11 bins, width = 0.352 Bq m^{-3} , first bin starting at 0.308 Bq m^{-3}) and (c) hour-of-day (11 bins, first bin starting at 09:00 UTC, time is colour-coded (Carmeri, 2018)). For (b) and (c) the equation of the regression line and the results of a Pearson correlation analysis are presented at the panel top. The length of the grey bars depicts the multiplicative standard deviation (Limpert et al., 2001). The circle areas are proportional to the number of samples per bin.

Flux densities estimated for RTM days based on the implementation of the traditional RTM (Sect. 2.4.1.) varied between $85 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$ to $1203 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$ (GLDAS-Noah v2.1) and $59 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$ to $834 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$ (ERA5-Land), respectively (Table 2, left). Although these ranges are wide, 70% of the flux densities were within a factor of three. Mean daily INP₋₁₀ flux densities modelled in the Bayesian framework (Sect. 2.4.2) were within a similar range as flux densities estimated using the traditional implementation of the RTM (Table 2, right). The median of the modelled daily flux densities was approximately 100% (GLDAS-Noah v2.1) and 190% (ERA5-Land) larger than medians obtained using the implementation of the traditional RTM.



Table 2: (left) Radii of the circular area centred around JFJ, mean radon exhalation rates within this area as calculated from two monthly mean radon flux maps (Karstens and Levin, 2023), slope m of a linear regression line between parallel INP₋₁₀ and radon-222 measurements and the resulting INP₋₁₀ flux density on RTM days calculated based on the traditional implementation of the RTM for both radon flux maps. (right) INP₋₁₀ flux density modelled in a Bayesian framework.

date	radius (km)	implementation of the traditional RTM				box model approach			
		radius (km)	mean radon exhalation (Bq m ⁻² h ⁻¹)		m	flux density (INP ₋₁₀ m ⁻² h ⁻¹)		flux density (INP ₋₁₀ m ⁻² h ⁻¹)	
			GLDAS-Noah	ERA5-Land		GLDAS-Noah	ERA5-Land	HDI 3%	HDI 97%
2022-07-02	43	81	81	56	1.05	85	59	30	267
2022-07-09	65	88	88	61	13.68	1203	834	440	1178
2022-07-14	81	91	91	64	5.41	492	346	565	1209
2022-07-18	81	91	91	64	3.92	357	251	323	723
2022-07-19	58	86	86	57	1.97	169	112	14	231
2022-08-03	65	93	93	66	4.43	412	293	376	1058
2022-08-13	97	98	98	72	2.24	220	162	27	300
Median of daily values						357	251		723
Median of values binned by hour of day		91		64	2.79	254	179		
Median of values binned by radon		91		64	1.83	167	117		



To our knowledge, the work presented here is the first application of the RTM to estimate flux densities of particles. It is therefore prudent to compare our results to particle fluxes measured with different approaches. Lindemann et al. (1982) derived on a particular summer day by the aerodynamic gradient method in the first few meters above an agricultural field an emission flux of culturable IN bacteria active ≥ -10 °C of around $4300 \text{ m}^{-2} \text{ h}^{-1}$. This is a 3 to 4 times greater value than the highest INP₋₁₀ flux values determined for the airshed around JFJ. However, Lindemann et al. (1982) had made their measurement close to a snap bean canopy “which supported a substantial population of epiphytic INA bacteria”.

A major fraction of airborne INPs is often super-micron in size (e.g. (Bras et al., 2024; Creamean et al., 2018; He et al., 2023; Lacher et al., 2026; Mason et al., 2016)). Therefore, we also estimated the flux of particles $> 2 \mu\text{m}$ by the same implementation of the traditional RTM, using the same mean radon exhalation rates and measurement intervals as for INP₋₁₀. The resulting mean flux densities were 1.0×10^8 (GLDAS-Noah v2.1) and 7.0×10^7 (ERA5-Land) particles $> 2 \mu\text{m} \text{ m}^{-2} \text{ d}^{-1}$. Employing the eddy covariance technique, Bignotti et al. (2022) had quantified aerosol fluxes above a deciduous forest located around 250 km southeast of JFJ. Their observed mean daily upward flux for measurements conducted between April and October was 2.9×10^7 particles $> 2 \mu\text{m} \text{ m}^{-2} \text{ d}^{-1}$ which is within a factor of three of our estimates derived from applying the implementation of the traditional RTM.

Retrospectively, the implementation of the traditional RTM can also be applied to estimate the flux density of INP₋₃₀ as reported by Brunner et al. (2022), who monitored INP₋₃₀ concentrations at JFJ over one year with a temporal resolution of 20 min. In their Fig. 6, the authors report median diurnal phase-averaged INP₋₃₀ and radon concentrations during a convective period between the end of March and mid-April, 2020. Reading the values at every full hour between 11:00 UTC and 16:00 UTC yields a slope (m) of $7182 \frac{\text{INP}_{-30}}{\text{Bq}}$ ($r = 0.98$), resulting in flux density estimates of $6.7 \times 10^5 \text{ m}^{-2} \text{ h}^{-1}$ (GLDAS-Noah v2.1) and $4.7 \times 10^5 \text{ m}^{-2} \text{ h}^{-1}$ (ERA5-Land), which is 2.6×10^3 times as large as our estimates for INP₋₁₀ flux densities that are also based on the median of values binned by hour of day. This difference in estimated flux densities is comparable to the observed difference in the number of atmospheric INP₋₁₀ and INP₋₃₀ (Kanji et al., 2017; Petters and Wright, 2015).

The flux density of INP₋₁₀ is likely subject to short-term and small-scale variation on timescales < 1 day (Hirano and Upper, 1989; Lindemann et al., 1982). The greater flux estimate for measurements binned by hour-of-day as compared with measurements binned by radon concentration (Fig. 5 (b) and (c), Table 2, left) might indicate an increase in INP₋₁₀ emissions in the source region between morning and afternoon. The sampled INP₋₁₀ population probably comprises an important fraction of INPs associated with biological material (Conen et al., 2022; Einbock and Conen, 2024; Huang et al., 2021). Some bacteria, fungal and pollen species were found to be emitted in diurnal cycles from different ecosystems (Drautz-Moses et al., 2022; Gusareva et al., 2019; Helbig et al., 2004; Hirst, 1953; Lindemann and Upper, 1985). Furthermore, increasing turbulence and wind speed between morning and afternoon on clear-sky summer days (Whiteman, 2000) might enhance regional scale



455 dispersal efficiency of emitted aerosols (Jones and Harrison, 2004). Moreover, higher flux estimates for measurements binned by hour-of-day could also result from greater INP_{-10} source activities with distance from the site.

3.1.2 Estimated contribution of thermally driven mountain winds to INP_{-10} export

Ice nucleating particles precipitating from clouds must have been transported to the corresponding height beforehand. Knowing the amount of INP_{-10} deposited by precipitation around JFJ and the frequency of thermally driven mountain winds allows to
 460 roughly estimate the contribution of the anabatic INP_{-10} flux to the overall amount of INP_{-10} reaching this high-altitude site. Zellweger et al. (2003) and Griffiths et al. (2014) investigated the frequency of thermally induced PBL intrusions at JFJ over the course of two years. Thermally induced vertical air transport occurred 34% of the time between March and August (Zellweger et al., 2003). Anabatic mountain winds reached JFJ on 40% of days between April and September (Griffiths et al., 2014). On average, precipitation at JFJ contains around 5 INP_{-10} mL (Stopelli et al., 2015) and the average annual precipitation
 465 in Switzerland is 1200 mm. This corresponds to a yearly INP_{-10} deposition in Switzerland of around $6 \times 10^6 \text{ INP}_{-10} \text{ m}^{-2}$. An anabatic upward flux of $120 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$ to $1210 \text{ INP}_{-10} \text{ m}^{-2} \text{ h}^{-1}$, the range of rounded median flux densities as obtained from the implementation of the traditional RTM (Table 2, left), and the Bayesian framework (Table 2, right), during 9 h (Henne et al., 2004) on 40% of days between April and September would only replace 1% to 13% of a total of 6×10^6 yearly by precipitation deposited $\text{INP}_{-10} \text{ m}^{-2}$ in the sink region. Although thermally driven mountain winds seem to play a distinctive role
 470 in shaping diurnal variations in INP_{-10} concentrations at JFJ during summer, this rough estimate could be a first clue that the majority of INP_{-10} deposited by precipitation in Switzerland is transported to cloud height during different meteorological conditions and by other mechanisms than thermally driven mountain winds. These mechanisms could for example include lifting by convective cells (Daily et al., 2026; Schaefer et al., 2026), föhn winds or frontal systems which have been shown to efficiently mix air parcels from a large catchment area above the usual height of the PBL (Jin et al., 2022). Locally, precipitation
 475 associated with and strong updrafts in thunderstorm cells might be another mechanism efficiently exporting biological INPs to the FT. Interestingly, thermally induced transport contributes distinctly to total aerosols found at JFJ (Lugauer et al., 1998; Zellweger et al., 2003). This potential difference in the importance of thermally driven mountain winds in contributing total aerosols and INP_{-10} to ambient particle populations at JFJ could be related to differences in emission behavior with varying meteorological conditions. Rainfall, for example, can lead to a significant increase in atmospheric biological INP
 480 concentrations while concurrently reducing total aerosol numbers (e.g. (Conen et al., 2017; Huffman et al., 2013; Mignani et al., 2021; Testa et al., 2021). The effect of rainfall on ambient biological INP abundance, for example, might reduce the importance of thermally driven mountain winds which prevail under clear sky conditions in transporting INP_{-10} to cloud height as compared to total aerosols and perhaps INP populations dominated by mineral dust, despite the frequency of PBL air injections caused by anabatic mountain winds.

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It could be interesting to investigate and compare INP flux densities during different meteorological conditions in the JFJ airshed and from fetch regions of varying sizes as well as for INPs active at different temperatures. Distinct PBL plume events at the site do not only occur during fair-weather conditions, but also during periods with strong synoptic scale winds or frontal passages (Zellweger et al., 2003). Implementing the RTM during such events might provide a possibility to quantify the effect of variations in relative humidity (Wright et al., 2014), precipitation or wind speed on the emission flux density of biological and other types of INPs across a source area that are transported to the site.

3.2 An exceptional case

Criteria b and c were not met on 08 July 2022, when Arctic air masses with no land contact for 10 days arrived at JFJ between 11:00 UTC and 13:00 UTC (Figure 6). Expectedly, they contained little radon (0.39 Bq m^{-3} and 0.47 Bq m^{-3}), but unexpectedly the highest INP concentrations found throughout the entire study ($43.0 \text{ INP}_{-10} \text{ m}^{-3}$ and $45.7 \text{ INP}_{-10} \text{ m}^{-3}$). Typically, few INPs populate the Arctic boundary layer because of weak local INP sources. However, neighbouring boreal regions are rich in INPs (Conen et al., 2023; Lill et al., 2024) and can raise INP concentration in High Arctic air (Porter et al., 2022), where the lower free troposphere sometimes carries enhanced aerosol loads (Igel et al., 2017). Correspondingly, the concentration of INP_{-10} in elevated Arctic aerosol layers occasionally exceeds 10^2 INP_{-10} (Raif et al., 2024). A further peculiarity of this exceptional case was the ratio of INP_{-10} to particles $\geq 0.5 \mu\text{m}$ (3.3×10^{-4} and 1.8×10^{-4}) that was well above the third largest such ratio found throughout the entire campaign (7.0×10^{-5}). Thus, this exceptional case probably resulted from air masses having been influenced by seemingly stronger INP but weaker radon sources (Conen and Robertson, 2002; Williams et al., 2009) as compared to the surroundings of JFJ. The different behaviour of the two atmospheric constituents during their long-distance transport may further have added to this unusual observation.

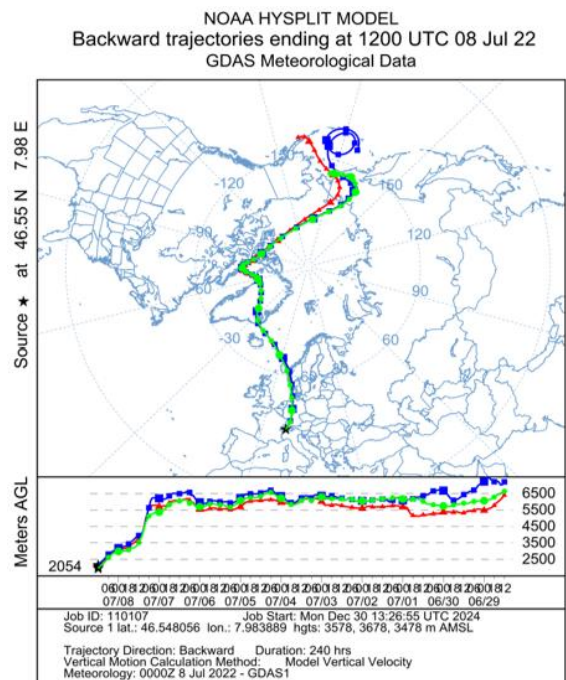


Figure 6: Backward-trajectories (240 h) of air parcels released at Jungfraujoch on 08 July 2022 at 12:00 UTC as calculated by the HYSPLIT model from GDAS1 meteorology. Release altitudes in the model were 3478 m, 3578 m, and 3678 m a. s. l. Temperature and relative humidity at 3578 m a. s. l. were identical in the model and in situ (3 °C, 12%).

4. Conclusions

To conclude, our results suggest that the RTM can be applied to estimate INP flux densities between terrestrial INP source regions and remote sites during periods influenced by PBL intrusions. The implementation of the RTM at high-altitude sites enables a direct quantification of the INP emission flux from a specific source area that reaches cloud height. Investigations at high-altitude as well as island sites might provide insights about INP emission flux densities from source areas comprising varying land cover and land use types. Long time series with a high temporal resolution of parallel INP and radon measurements at remote sites might provide an opportunity to compare emission fluxes during different seasons and meteorological conditions in the INP source area. Large datasets make it possible to filter events fulfilling all prerequisites for the implementation of the RTM without precluding extensive analysis and interpretation. An interesting investigation would further be the comparison of INP emission flux densities estimated by the RTM with bottom up approaches. Finally, flux densities from various source regions and during different meteorological conditions might be used to inform the representation of INP dynamics in weather and climate models.



520 Appendix A

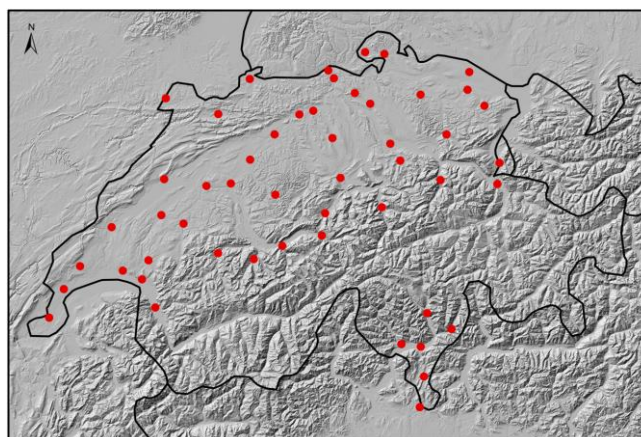


Figure A1: Locations of the meteorological stations operated by the Swiss Federal Office of Meteorology and Climatology (MeteoSwiss) from which data on daily total sunshine duration, daily global radiation and wind speed were obtained. Data source relief map: Swiss Federal Office of Topography (swisstopo).

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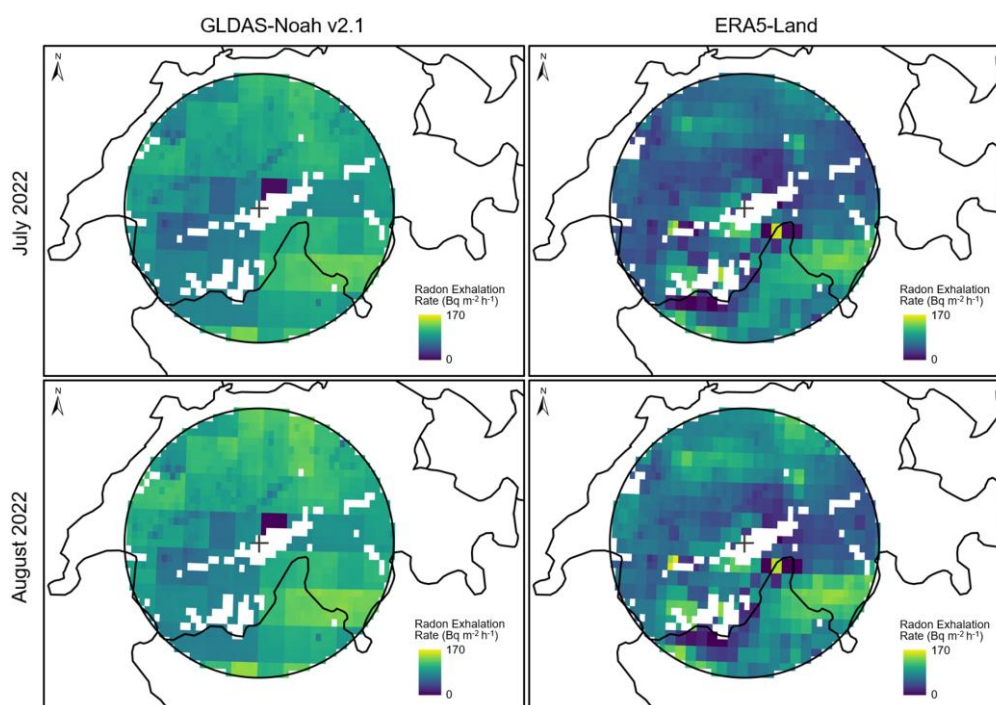


Figure A2: Mean monthly radon exhalation rates obtained from two process-based European radon flux maps available at the ICOS Carbon Portal (Karstens and Levin, 2023) based on either the GLDAS-Noah v2.1 (Karstens and Levin, 2024a, left) or the ERA5-Land (Karstens and Levin, 2024b, right) soil moisture reanalysis dataset for July (top) and August (bottom), 2022, within a radius of 100 km from the Jungfrauoch Observatory (black cross).

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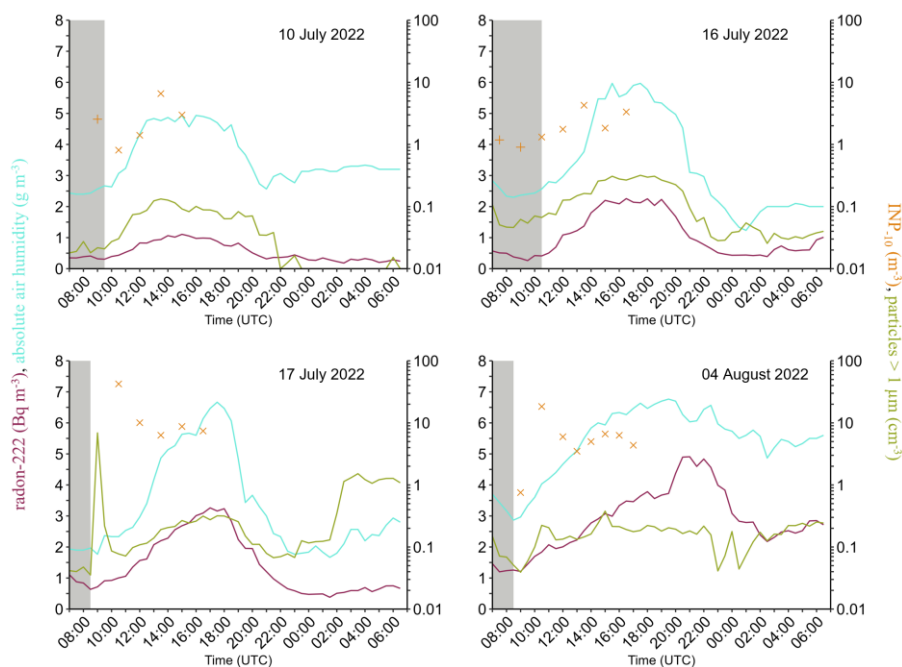


Figure A3: Temporal development of the radon concentration (purple), absolute air humidity (blue) and number of particles $> 1 \mu\text{m}$ (green) for days fulfilling all but validation criteria b. Orange crosses and saltire display INP_{10} concentrations before and after the onset of a thermally driven radon increase respectively. Grey shaded area corresponds to the time before the thermally driven increase in radon concentrations.



Appendix B

Table B1: Cumulative concentration of ice nucleating particles active at temperature T, K(T), measured at Jungfraujoch and corrected for potential INP loss during sample correction by multiplying uncorrected INP concentrations with $\frac{2}{1+0.7}$. *Italic font indicates extrapolated concentrations.*

Date	Time (UTC)	K(T) (m ⁻³)														
		-3	-3.5	-4	-4.5	-5	-5.5	-6	-6.5	-7	-7.5	-8	-8.5	-9	-9.5	-10
2022-07-01	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.56	0.95	1.57	2.01	2.24	2.71	4.57
2022-07-01	12:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.34	0.69	0.69	1.44	1.84	2.71	3.67
2022-07-01	14:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.91	1.11	1.30	2.37	3.07	4.10
2022-07-01	15:30	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.18	0.36	0.74	1.33	1.54	1.54	2.19	2.65
2022-07-01	17:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.17	0.35	0.70	1.08	1.47	1.47	2.77
2022-07-02	7:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.37	0.56	0.56	0.95	1.15
2022-07-02	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.17	0.17	0.34	1.06	1.06	1.44	1.64	2.26	2.94
2022-07-02	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.17	0.17	0.34	0.34	0.69	0.69	0.87
2022-07-02	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.18	0.18	0.35	0.72	0.91
2022-07-02	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.75	1.15	1.79	2.24	2.71	3.46	4.57
2022-07-02	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.38	0.58	0.99	0.99	1.20	1.42	1.42	2.82
2022-07-02	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.58	0.78	1.42	1.86	2.33	2.82	3.61
2022-07-08	11:30	0.00	0.00	0.00	0.00	0.17	0.17	0.72	2.35	6.20	9.49	17.93	25.51	35.33	35.33	43.01
2022-07-08	13:00	0.00	0.00	0.00	0.00	0.00	0.37	0.76	0.96	2.49	6.22	9.07	16.65	27.07	37.50	45.65
2022-07-08	14:30	0.00	0.00	0.00	0.00	0.00	0.00	0.76	1.58	2.15	5.47	7.96	10.49	13.65	16.88	18.86
2022-07-08	16:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.76	0.76	1.56	2.13	3.35	7.45	7.45	8.36
2022-07-09	8:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.38	0.58	0.78	0.98	1.41	2.08	3.58	5.03
2022-07-09	9:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.57	1.38	2.04	2.75	3.79	5.59	6.27
2022-07-09	11:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.58	0.78	1.42	1.64	1.86	2.09
2022-07-09	12:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.84	1.06	1.29	1.52	2.00	3.02	3.58
2022-07-09	14:00	0.00	0.00	0.00	0.00	0.00	0.00	0.57	0.97	1.60	2.04	2.27	3.52	3.79	4.65	5.59
2022-07-09	15:30	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.82	2.18	3.75	5.61	10.82	14.95	17.88	19.09
2022-07-09	17:00	0.00	0.00	0.00	0.00	0.00	0.00	0.41	1.52	2.50	3.86	5.44	7.28	8.57	9.53	11.15
2022-07-10	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.19	0.58	0.58	1.19	1.41	2.55
2022-07-10	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.40	0.60	0.81	0.81
2022-07-10	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.58	0.78	0.98	1.41
2022-07-10	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.80	1.01	1.45	1.91	3.97	4.57	6.58
2022-07-10	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.62	1.05	1.27	2.21	2.46	2.98
2022-07-14	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.37	0.75	0.95	0.95	0.95
2022-07-14	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.78	1.19	1.41	1.85	1.85	2.31	3.85	4.42
2022-07-14	13:30	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.58	0.78	1.42	1.42	1.64	2.09	2.33	2.57



2022-07-14	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.83	1.28	2.48	4.43	5.40	5.40	7.23	10.50
2022-07-14	16:30	0.00	0.00	0.00	0.00	0.20	0.62	0.84	3.02	4.47	6.51	9.04	10.04	11.15	14.57	16.33
2022-07-14	18:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	1.06	2.76	3.02	4.78	6.89	9.04	10.58
2022-07-15	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.98	1.62	1.85	3.31	4.13	5.35
2022-07-15	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.61	0.82	1.03	1.25	1.94	2.93	4.04
2022-07-15	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.61	1.25	2.42	3.20	4.33	5.27	6.31
2022-07-15	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.58	0.78	1.41	2.31	3.31	3.85	3.85	5.35
2022-07-15	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.61	0.82	2.18	2.68	3.75	5.27	5.27	5.95
2022-07-15	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	1.04	2.44	3.78	3.78	4.99	6.73
2022-07-16	7:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.19	0.38	0.57	0.57	1.18
2022-07-16	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.53	0.72	0.72	0.72	0.90
2022-07-16	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.54	0.73	0.92	1.12	1.32
2022-07-16	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.36	0.55	0.55	0.74	1.13	1.34	1.34	1.55	1.76
2022-07-16	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.75	0.95	1.36	2.47	2.95	4.28
2022-07-16	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.98	0.98	0.98	0.98	1.39	1.39	1.83
2022-07-16	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.58	1.20	1.86	2.57	3.08	3.34
2022-07-17	10:30	0.00	0.00	0.00	0.00	0.00	1.72	3.09	7.45	10.85	15.37	18.97	25.11	34.85	34.85	42.42
2022-07-17	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.37	1.37	4.03	5.54	5.87	6.95	8.16	10.07
2022-07-17	13:30	0.00	0.00	0.00	0.00	0.00	0.19	0.38	1.18	1.83	2.06	2.78	3.28	4.68	5.31	6.32
2022-07-17	15:00	0.00	0.00	0.00	0.00	0.00	0.38	0.38	0.98	2.06	2.53	3.55	4.10	5.97	7.07	8.75
2022-07-17	16:30	0.00	0.00	0.00	0.00	0.00	0.20	0.40	1.02	1.02	1.69	2.40	4.01	5.57	5.57	7.41
2022-07-18	9:00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.19	0.39	0.79	1.21	1.43	2.11	2.84	3.36
2022-07-18	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.39	0.80
2022-07-18	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.58	0.98	1.19	2.55	3.58
2022-07-18	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.38	0.98	1.83	2.06	2.53	2.78
2022-07-18	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.40	1.25	1.94	2.18	3.47	5.27	8.77
2022-07-18	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.55	1.13	1.33	2.64	2.88	3.37	4.17	6.01	7.10
2022-07-19	9:00	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.18	0.36	0.54	1.12	1.12	1.32	1.52	1.52
2022-07-19	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.38	2.04	2.75	3.52	4.95	5.92	7.01	7.81
2022-07-19	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.35	0.35	0.91	1.30	1.93	3.07	3.57	4.96	5.91
2022-07-19	14:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.36	0.73	1.33	1.96	3.90	4.17	7.10	7.89
2022-07-19	15:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.39	0.80	1.68	2.14	3.97	4.87	5.19	7.35
2022-07-19	17:00	0.00	0.00	0.00	0.00	0.00	0.00	0.78	0.99	0.99	1.86	2.57	3.34	4.46	6.43	8.89
2022-07-26	7:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.59	0.79	1.00	1.43	1.65	2.35	2.84
2022-07-26	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.38	0.38	0.38	0.38	0.58	0.98
2022-07-26	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.38	1.61	2.29	2.53	2.78	3.03
2022-07-26	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.56	0.75	1.15	1.57	1.79
2022-07-26	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.37	0.94	1.14	1.77	3.18	3.97
2022-07-26	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.19	0.97	0.97	1.17	1.38	2.27	3.79
2022-07-26	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.75	0.95	0.95	1.79	2.47	3.20	4.28	6.17	8.53
2022-07-27	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.61	1.03	1.25	1.25	1.25	2.42	3.47



2022-07-27	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.40	0.82	1.48	1.71	2.19	3.50
2022-07-27	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.56	0.56	0.56	0.56	0.76	0.76	1.16	1.37	2.49
2022-07-27	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.57	0.98	1.61	2.29	3.55	5.63	5.97	7.87
2022-07-27	15:00	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.99	1.86	2.82	3.88	4.76	5.39	6.43	8.43
2022-07-27	16:30	0.00	0.00	0.00	0.00	0.19	0.19	0.78	1.20	2.09	2.57	3.61	4.17	5.39	6.07	6.80
2022-07-27	18:00	0.00	0.00	0.00	0.00	0.00	0.18	0.18	0.36	0.94	1.55	2.90	3.15	4.79	5.09	6.42
2022-07-28	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.58	1.19	2.80	4.42	7.12	9.29	9.79	10.32
2022-07-28	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.77	0.98	1.39	2.53	3.28	3.82	4.68	5.31	7.46
2022-07-28	12:00	0.00	0.00	0.00	0.00	0.00	0.39	0.39	1.00	1.00	1.43	2.35	3.10	3.36	3.36	4.49
2022-07-28	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.01	1.91	3.15	4.57	5.86	6.21	7.35
2022-07-29	6:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.39	0.39	0.59	0.80	1.01	1.89	1.89
2022-07-29	8:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.38	0.78	1.42	1.86	1.86	2.09	2.57
2022-08-03	10:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.17	0.35	0.53	0.90	1.28	1.68
2022-08-03	11:30	0.00	0.00	0.00	0.00	0.00	0.00	0.37	0.56	0.75	1.57	2.01	2.71	2.95	3.73	5.83
2022-08-03	13:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.56	0.75	1.57	2.95	4.87	5.50	7.68
2022-08-03	14:30	0.00	0.00	0.00	0.00	0.00	0.38	0.58	1.86	3.34	5.39	8.00	9.37	10.40	12.19	14.38
2022-08-03	16:00	0.00	0.00	0.00	0.00	0.00	0.00	0.64	0.86	2.04	2.29	3.37	3.66	4.25	5.91	7.87
2022-08-03	17:30	0.00	0.00	0.00	0.00	0.00	0.40	0.82	2.44	3.22	3.78	6.36	9.32	13.48	15.07	15.96
2022-08-03	19:00	0.00	0.00	0.00	0.00	0.42	0.42	0.86	1.09	2.29	3.95	4.89	5.91	6.65	8.31	9.24
2022-08-04	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.37	0.37	0.37	0.37	0.56	0.75
2022-08-04	10:30	0.00	0.00	0.00	0.00	0.19	0.38	0.99	3.34	3.88	7.18	8.43	11.56	15.23	17.20	18.35
2022-08-04	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.38	1.18	1.83	2.06	2.53	4.10	4.99	5.97
2022-08-04	13:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.56	0.56	0.56	1.16	1.80	2.73	3.49	3.49
2022-08-04	14:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.38	1.18	2.06	3.03	4.10	4.99
2022-08-04	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	1.24	1.92	2.40	3.44	4.01	4.60	5.57	6.63
2022-08-04	16:00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	1.03	1.25	1.94	2.68	2.93	4.64	5.27	6.31
2022-08-04	17:00	0.00	0.00	0.00	0.20	0.20	0.40	0.61	0.61	1.04	1.49	1.95	2.44	3.22	4.07	4.37
2022-08-05	8:30	0.00	0.00	0.00	0.00	0.20	0.20	0.40	0.81	1.46	4.01	4.01	4.60	4.91	5.23	7.01
2022-08-05	10:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.58	1.20	1.42	1.42	1.86	3.61
2022-08-05	11:30	0.00	0.00	0.00	0.00	0.00	0.00	0.30	0.30	1.25	1.58	2.98	4.51	5.76	7.61	8.62
2022-08-05	13:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.29	0.58	2.46	2.80	3.87	5.02	5.42	6.26
2022-08-06	7:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.41	0.62	0.62	0.83	1.05	1.05	1.27
2022-08-06	9:30	0.00	0.00	0.00	0.00	0.00	0.00	0.28	0.28	0.57	0.57	0.87	0.87	0.87	0.87	0.87
2022-08-06	11:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.27	0.55	0.55	0.55	0.55	1.12	1.12
2022-08-06	12:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.62	1.50	1.97	2.21	2.21	2.72	2.98	3.81
2022-08-06	14:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.60	0.81	1.46	1.69	2.65	3.44
2022-08-06	15:30	0.00	0.00	0.00	0.00	0.00	0.39	0.39	0.39	1.00	1.00	1.88	2.35	2.84	3.63	4.80
2022-08-06	17:00	0.00	0.00	0.00	0.00	0.00	0.00	0.43	0.65	1.10	1.34	1.58	1.58	2.33	4.32	6.00
2022-08-07	9:00	0.00	0.57	0.57	0.57	0.57	0.77	0.77	0.77	0.77	0.77	0.97	0.97	1.38	2.51	3.79
2022-08-07	10:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.60	0.81	1.02	1.46	1.69	1.69	2.16
2022-08-07	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.40	0.61	1.04	2.19	2.44	2.44	2.96



2022-08-12	9:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.41	0.62	1.50	2.46	3.25	3.52	4.10	4.71
2022-08-12	11:00	0.00	0.00	0.00	0.00	0.00	0.20	0.20	0.40	0.82	1.49	2.44	3.50	6.36	7.12	9.82
2022-08-12	12:30	0.00	0.00	0.00	0.00	0.20	0.41	0.41	0.41	0.41	0.83	1.74	3.00	5.06	5.74	7.23
2022-08-12	15:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.62	1.51	4.43	6.09	8.97	11.07	13.69	17.20
2022-08-12	17:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.39	0.59	1.43	2.11	2.84	3.36	4.49	5.77
2022-08-13	9:00	0.00	0.00	0.00	0.00	0.00	0.00	0.55	0.75	1.14	1.14	1.14	1.35	1.56	1.77	2.45
2022-08-13	10:30	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.19	0.78	0.99	1.20	1.64	2.33	3.08	4.17
2022-08-13	12:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.17	0.72	2.13	2.35	4.07	5.22	6.20	6.20
2022-08-13	13:30	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.39	0.59	0.80	2.37	2.37	2.87	4.23	5.15
2022-08-13	15:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.82	0.82	1.04	1.95	2.44	3.78	4.07
2022-08-13	16:30	0.00	0.00	0.00	0.00	0.00	0.00	0.40	1.24	1.69	2.40	5.57	7.01	7.41	7.82	7.82
2022-08-13	18:00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.40	0.61	1.70	2.68	3.20	4.33	5.95	7.07
2022-08-14	8:30	0.00	0.00	0.00	0.00	0.00	0.19	0.19	0.60	1.68	2.14	4.27	6.21	7.35	8.19	9.10
2022-08-14	10:00	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.39	0.80	1.23	2.63	3.15	4.87	6.21	6.96
2022-08-14	11:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.38	1.20	1.86	3.34	3.61	6.07	8.00	9.37
2022-08-14	13:00	0.00	0.00	0.00	0.00	0.00	0.21	0.65	1.59	2.61	3.16	4.67	5.68	6.80	8.49	11.65
2022-08-14	14:30	0.00	0.00	0.00	0.00	0.00	0.20	0.40	0.81	1.69	2.65	4.01	5.23	7.01	8.70	10.19
2022-08-14	16:00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.60	0.81	2.16	3.44	4.91	5.57	7.82	9.17
2022-08-14	17:30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.79	1.21	2.35	3.10	4.49	4.80	6.12	6.85

Code and data availability

The INP dataset is provided in Appendix B. The box model code will be publicly accessible on zenodo after publication of the manuscript (<https://doi.org/10.5281/zenodo.23411550>). Aerosol data is available upon request.

545 Author contributions

AE, FC, ADG and SDC conceptualized the study. AE collected and analysed the INP samples and visualized the data. ADG developed the box model and carried out the simulations with contributions from AE, FC and SDC. All authors analysed the data, wrote the original draft and reviewed and edited the manuscript.

Competing interests

550 The authors declare that they have no conflict of interest.



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