

INTRODUCTION

Ice nucleating particles (INPs) are essential for the formation of ice crystals in clouds, which affect the climate system and precipitation. Previous studies by Mossop (1966) and Stevenson (1967) have studied how measured INP concentration is affected by the sampled volume of air. We investigated the Volume-Concentration relationship of INPs at -15 °C. We found that the theoretical relationship did not always hold, and the reported concentration was not consistent when different volumes of air were sampled. Additionally, a longer time series analysis will be presented.

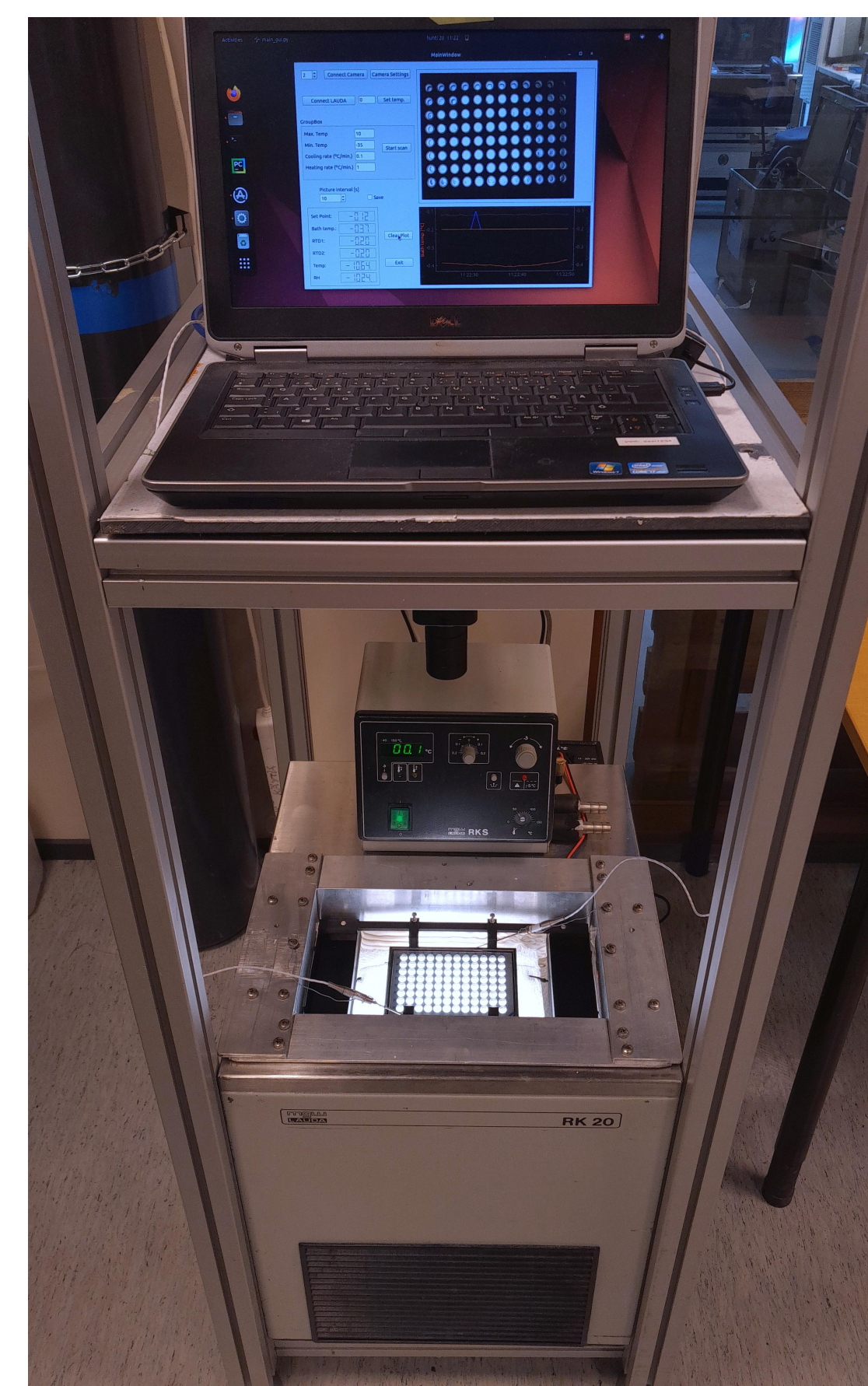
MATERIALS AND METHODS

FrESH instrument

FrESH (Freezing Experiment Setup Helsinki) is an experimental setup used to measure the concentration of ice nucleating particles (INP) in collected atmospheric aerosols, specifically in the immersion freezing mode. It consists of a camera, temperature-controlled ethanol bath, temperature sensors, and a computer with software to control and record data.

To analyze the collected atmospheric aerosols, an aqueous suspension of the particles is prepared, and a 96-well PCR plate is filled with the suspension. The plate is placed on the chiller, and the freezing process is monitored by taking pictures of the droplets at regular intervals.

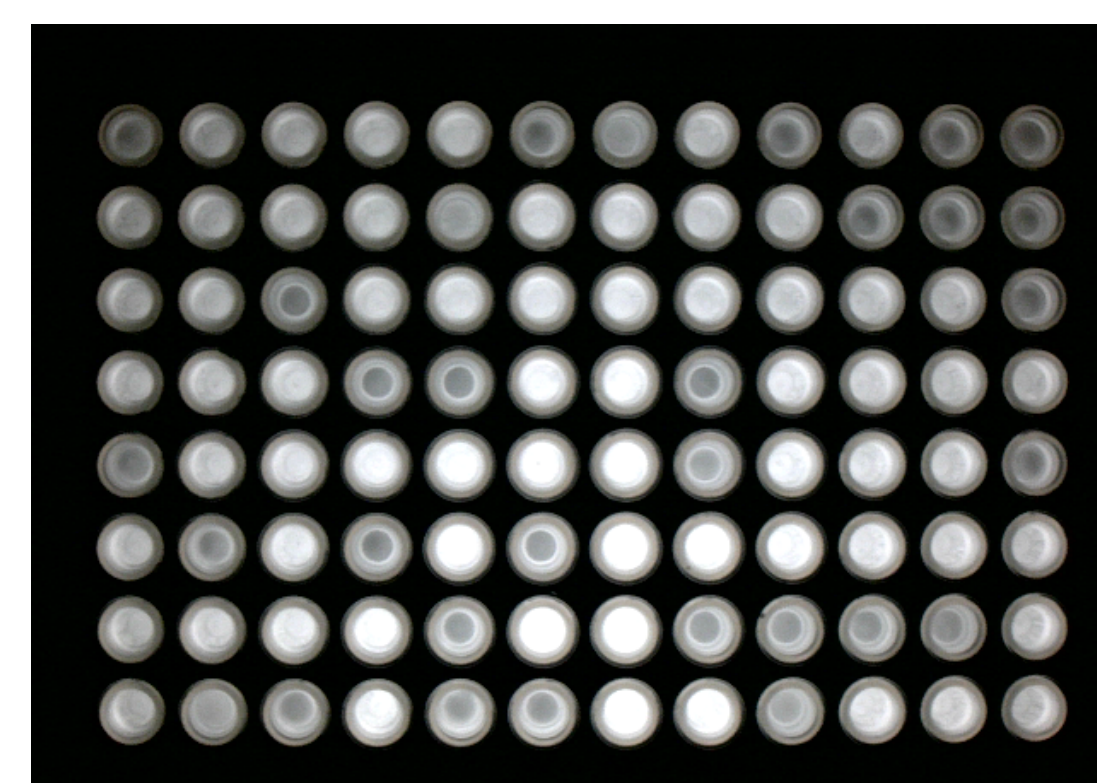
The frozen fraction of droplets as a function of temperature is calculated using the raw images, and converted to INP concentration using the volume of sampled air and the volume of the droplets, assuming a Poisson process (Vali 2019).



(a) FrESH instrument



(b) Filter inlets



(c) Example of a freezing experiment with some of the wells frozen

Description of the collection method

We used three automatic samplers to collect atmospheric samples in Helsinki. Each sampler collected atmospheric particles using 3 different sampling rates during 24 hours using 47 mm Whatman nuclepore membrane filters with a pore size of 0.2 μm. The filters are stored at -20°C until analysis.

RESULTS

Sampled volume - INP concentration relationship

To study the relationship between the measured INP concentration the volume of air sampled, we collected three filters per day simultaneously at three different sampling rates: 5.5 L/min, 16.6 L/min, and 33.3 L/min, with volumes ranging from 7000 to 50000 L over a period of 14 days. After discarding some faulty samples, we obtained a total of 35 filters.

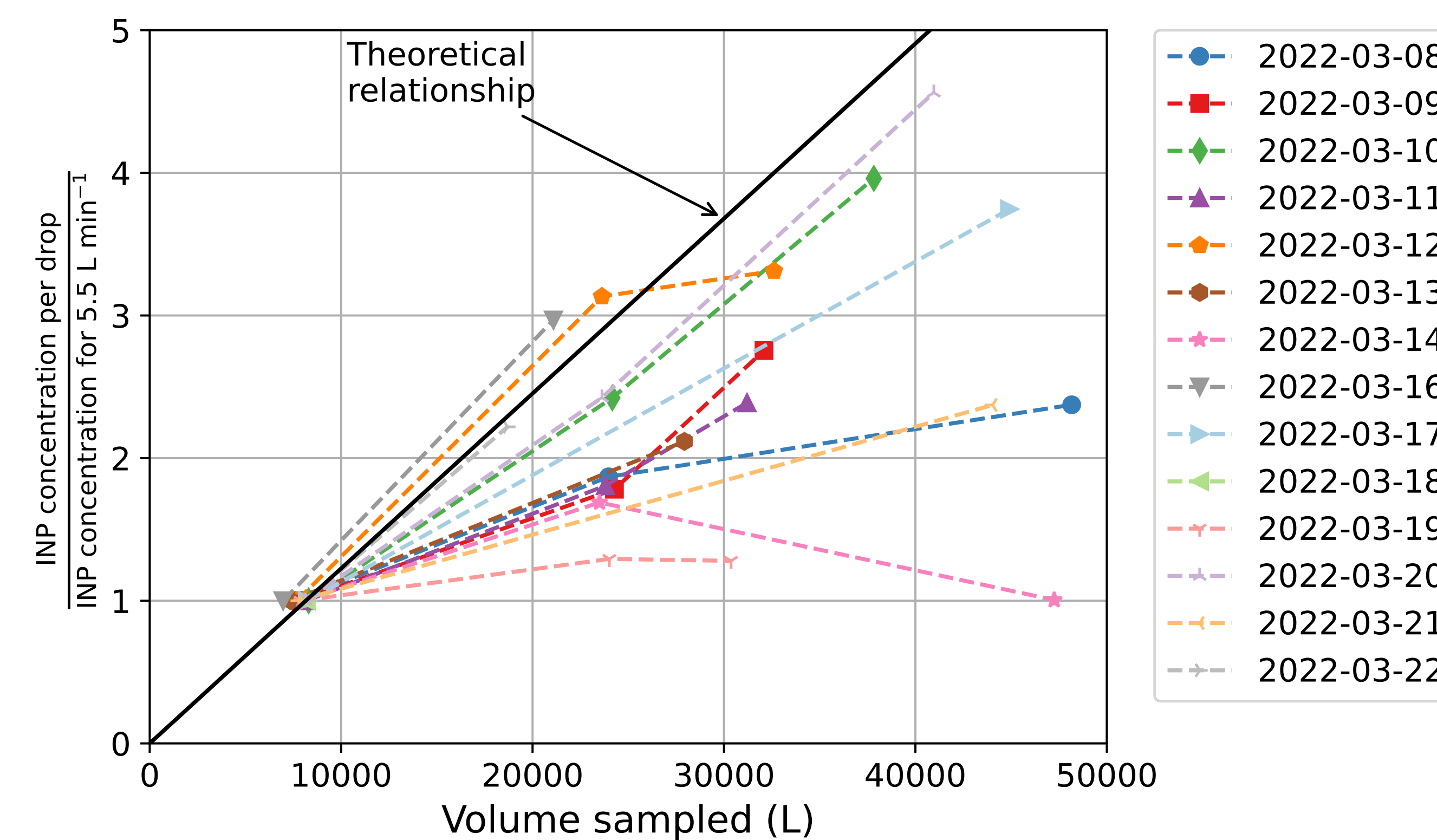


Figure 2. Volume-Concentration Relationship of INPs at -15 °C.

Figure 2 shows the Volume-Concentration Relationship of INPs at -15 °C. The black line in the figure represents the theoretical relationship, where a doubling of sampled air volume would result in a doubling of INP concentration per drop. However, we observed a range of behaviors in our results:

- Some observations closely followed the theoretical relationship.
- In other cases, the measured INP concentration remained constant even as the sampled air volume increased.
- This trend was observed across all measured temperatures.

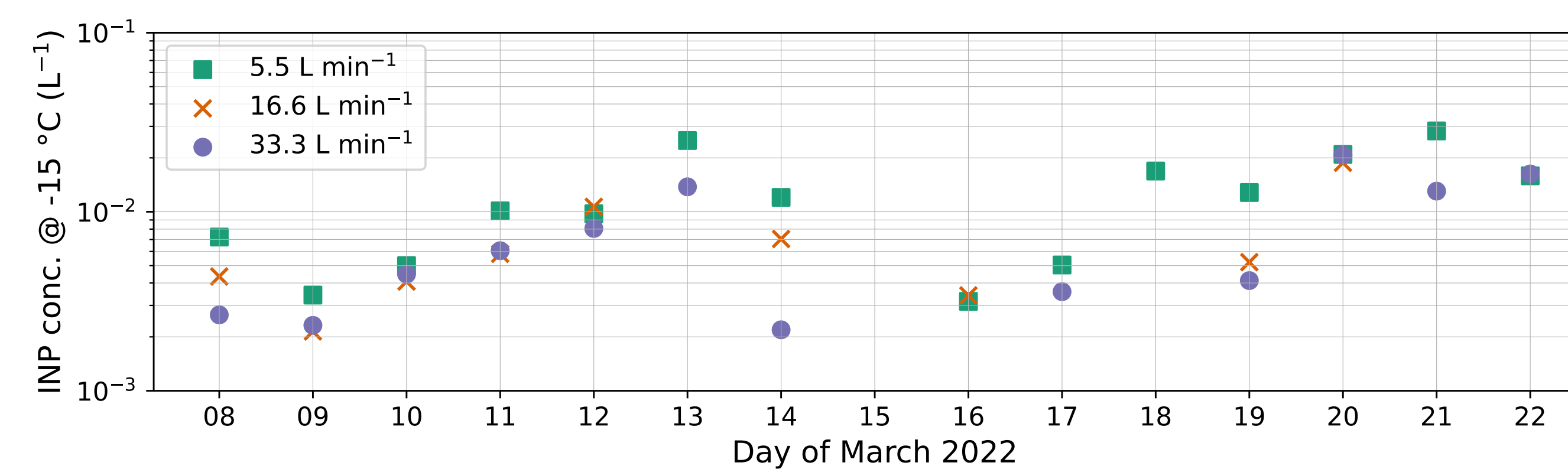


Figure 3. Volume-Concentration Relationship of INPs at -15 °C.

Figure 3 displays the time series of INP concentration per standard liter at -15 °C for the three different sampling rates. It is evident that the reported concentration is not consistent when different sampling rates are used. While the concentration remained the same on several days, a significant difference of almost one order of magnitude was observed on March 14. What remains consistent is that the reported concentration increases as the flow rate decreases.

Time series of INP concentration

To further investigate the variability of INP concentrations over time, we analyzed a longer time period from the sampler with a flow rate of 16.6 L/min. The time series of INP concentration at -15 °C is shown in Figure 4. Two distinct periods are shown: November 22, 2021 to December 24, 2021, and February 16, 2022 to March 30, 2022.

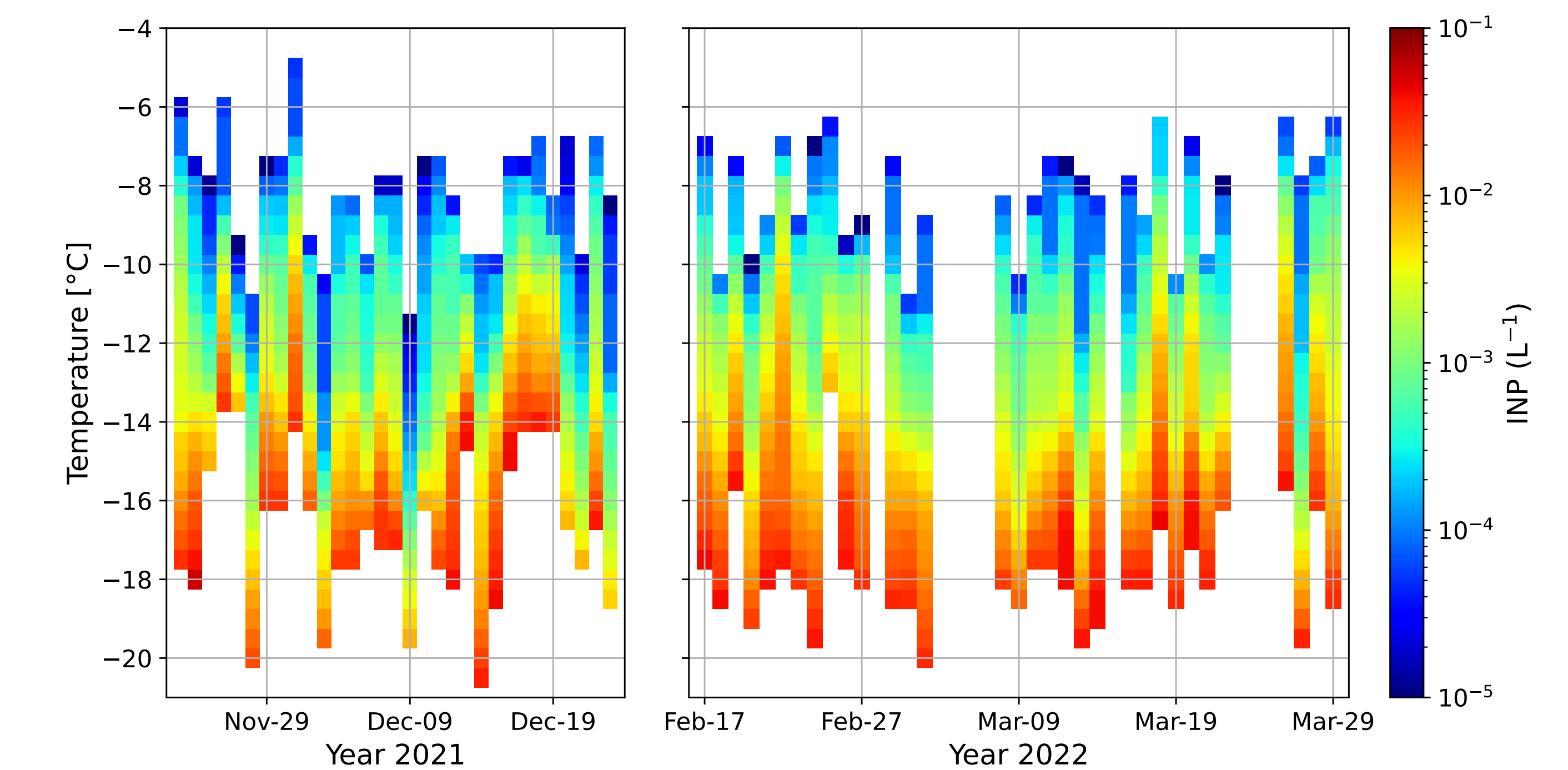


Figure 4. Time series of INP concentration.

The concentration exhibited a smooth behavior, with occasional jumps on certain days, which may be attributed to changes in local sources. The onset temperature of the experiments ranged between -5 to -11 °C.

CONCLUSIONS

This study examined ice nucleating particle (INP) concentration in the atmosphere using the FrESH assay. The Volume-Concentration Relationship of INPs at -15 °C showed that the theoretical relationship did not always hold. INP concentration was not consistent with different sampling rates, and decreased as flow rate increased. Long-term analysis showed a smooth concentration behavior with occasional changes attributed to local sources. INP concentrations observed are relevant to cloud formation at low temperatures (-5 to -11 °C). These findings aid understanding of climate system processes and predicting cloud formation and precipitation. Future studies could investigate other sampling parameters and sources of INP.

ACKNOWLEDGEMENTS

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References

- Mossop, S., (1966). The Use of Membrane Filters in Measurements of Ice Nucleus Concentration. I. Effect of Sampled Air Volume. *Journal of Applied Meteorology*, 5(4), 474-480. [https://doi.org/10.1175/1520-0450\(1966\)005<0474:TUOMFI>2.0.CO;2](https://doi.org/10.1175/1520-0450(1966)005<0474:TUOMFI>2.0.CO;2)
- Stevenson, C., (1967). An improved Millipore filter technique for measuring the concentrations of freezing nuclei in the atmosphere. *Quarterly Journal of the Royal Meteorological Society*, 94(399), 34-43. <https://doi.org/10.1002/qj.49709439905>
- Vali, G., (2019). Revisiting the differential freezing nucleus spectra derived from drop-freezing experiments: Methods of calculation, applications, and confidence limits. *Atmos. Meas. Tech.*, 12(2), 1219-1231. <https://doi.org/10.5194/amt-12-1219-2019>