We thank the Referees for their constructive comments on our paper and for finding our manuscript interesting for publication. Their suggestions helped us to elaborate more clearly the issues presented in the manuscript and to improve the quality of the work. Here is a point-by-point reply to their proposed corrections. We merged the reply to the Referees into a unique file to improve its clarity and the level of detail.

Referees' comments are in italic font, authors' response in blue normal font, changes in the manuscript are in blue underlined and refer to the document published on ACPD. To facilitate the discussion, we have numbered the comments.

Referee #1:

Stopelli and co-authors present a series of ice nucleating particle concentrations at -8 °C in precipitation ($INPs_{-8}$) from over one hundred samples taken at the Jungfraujoch Research station in Switzerland. The results, which show concentrations ranging from as high as 434 $INPs_{-8}$ ml^{-1} to below the limit of detection, are then compared with the range of 'standard' meteorological, aerosol and chemical observations at this site. The results from the first 80 % of the observations are then used to create models to predict the INP concentration, with the last 20 % of observations used as a model validation. The models provided suggest that the INP concentration in precipitation is potentially driven by wind speed, temperature, aerosol concentration, the season and source region, or the precipitation history of the air mass.

The manuscript is in my opinion ideal for publication in Atmospheric Chemistry and Physics, and is also in a good condition. I have a number of minor comments and some technical corrections (listed below), however it must be noted that based on my personal background an in-depth critique of the statistical model part of the paper cannot be given.

Minor Comments:

(1) *Introduction: The introduction explains the importance of INPs for cloud processes and precipitation formation and then talks about measurements of INPs and why they are relevant at high temperatures. However, I missed any discussion of how the observations made, specifically of INPs in collected precipitation water, relates to the cloud processes themselves. There are various processes that would increase or reduce the concentration of INP's in collected water; these and their possible effects on the measurements should be discussed.*

The connection between precipitation and cloud processes is particularly important and merits to be discussed in the manuscript. Impaction scavenging of aerosols can contribute to the removal of airborne INPs, increasing their abundance in precipitation. Crystal growth by vapour deposition and riming (if rime does not contain INPs) can on the contrary dilute INPs in precipitation. A possible improvement for similar top-down studies would be to couple measurements of airborne $INPs_{-8}$ with their abundance in precipitation and to find proxies for assessing the scavenging and riming efficiency. Concerning this manuscript, it has to be kept in mind that we were inside precipitating clouds, therefore as close as possible to the formation of precipitation. Furthermore, we generally mention $INPs_{-8}$ as "removed by precipitation" (coherently with Stopelli et al., 2015), which includes both mechanisms of active (nucleation) and passive (scavenging) removal. On page 6, line 37 we also indicate that important missing information is constituted by the "average mass (equivalent liquid volume) of hydrometeors formed by individual INPs". We dedicated a new paragraph to better clarify this issue at the end of section 2.1: "The collection of precipitation allows the sampling of INPs which either formed precipitating ice particles or were scavenged by precipitation. It is

difficult to distinguish between these contributions in field studies, where scavenging, riming and crystal growth by vapour deposition can alter the abundance of INPs in precipitation. Nevertheless, the station was always inside clouds while sampling, allowing us to collect falling snow as close as possible to where it formed. Furthermore, precipitation was immediately analysed, in order to minimise the chance for biases due to artefacts like the production (f.i. the release in solution of INPs or cellular multiplication) and the loss (f.i. settling or increased molecular weakness of biological INPs detached from mineral and soil dust) of $INPs_{-8}$".

(2) *Section 2.1.: How were the settling of particles and losses/experimental artefacts prevented/accounted for? What is the instrumental background?*

A detailed description of the blank preparation is provided in comment 1 Referee #2 and has been inserted in the manuscript at the end of section 2.1. Briefly, out of 29 blanks analysed only 2 had measurable $INPs_{-8}$ in concentrations of $0.11 \cdot ml^{-1}$ (the description of the instrument is fully reported by Stopelli et al., 2014). Settling of particles was prevented by gently shaking the sample prior to dividing it rapidly into aliquots. INPs active at moderate supercooling can increase or decrease, specifically when in solution, either due to cellular multiplication, release in solution of molecules, settling or even increased molecular weakness when biological residues detach from soil. We think that such behavior cannot be easily predicted and is sample-specific. The important aspect of this study is that we analysed the sample as soon as possible, trying to reduce as much as possible the impact any of the above mentioned artefacts. This is addressed at the end of section 2.1 in the revised version of the manuscript when discussing about sampling precipitation according to comment 1 Referee #1.

(3) *Section 2.1.: Is there any other details of the precipitation water? For example, particle concentration and size distributions could some indication (rather approximate) of the number of primary ice particles per ml of water. Some correlation with the concentration of particles could be expected.*

We do not have such information. Particle numbers in precipitation could be determined by flow cytometry. In practice, counting would probably be limited to larger particles. It could be approximate because INPs active at moderate supercooling can be constituted of cells, but also of cellular fragments and molecules, either freely floating or attached to soil and mineral dust particles of several dimensions. Furthermore, the number of particles in precipitation may be biased by the same scavenging/riming processes as it can happen for INPs, as discussed in comment 1 Referee #1.

(4) *Section 2.2.: This section launches straight in to a listing of the considered parameters. Some short introduction might be helpful to give some context.*

At the beginning of section 2.2 this sentence has been added to provide some context: "To analyse and understand more on the factors responsible for the variability of $INPs_{-8}$ in precipitation, several environmental parameters were considered in relation with the number of $INPs_{-8}$".

(5) *Section 2.2.: For the "source region score" and "season score", the text (specifically page 4 line 20) implies that numbers other than 1, 2, 3 and 4 were also considered. If so, it might be worth explicitly stating so and also stating the range of numbers considered.*

Yes, based on the *a priori* ranking of seasons and source regions several combinations of numbers were considered. To state it better, line 10 page 4 was changed into: "Therefore *a priori* the larger value should be assigned to events from South, followed by mixed conditions and by events from Northern Europe. Several combinations of values ranging from 1 to 3 (f.i. 2-1.5-1; 3-2-1,…) were tested and the best combination of values was determined through comparisons with the numbers of $\log(INPs_{-8} \cdot ml^{-1})$. It corresponds to: South = 3; mixed condition = 2, North = 1". Similarly, line 20 page 4 was changed into: "Once this *a priori* classification was established, the precise values for each class were again determined by comparing different possible combinations of numbers from 1 to 9 (f.i. 3-2-2-1; 9-6-3-1; 4-3-2-1,…) with measured values of $\log(INPs_{-8} \cdot ml^{-1})$. The best fit with the data was found for the combination: summer = 4; autumn = 3; spring = 2; winter = 1."

(6) *Section 2.3.: Does the exclusion of 'zero' samples not cause a bias towards higher values? Did you consider trying the same fitting process with, for example, random concentrations 0 <> 0.21 $INPs_{-8}$ ml^{-1}?*

Deciding to exclude the "zero" samples from the fitting process results in a poor constraint for the model at low concentrations, where error associated to counting frozen tubes is larger, but not necessarily in a bias towards larger values. The difference it introduces is that values above the detection limit have a weight in the fitting process and in evaluating the performance of a model. On the contrary, giving "zero" samples a value by randomly assigning a small concentration would have required a subjective judgement. Specifically, we would have had to subjectively decide on the shape of the random distribution >0 <0.21, which on a log scale can be relevant. To avoid any doubts about the objectivity in the model building process, we decided to accept the poor constraint at the very low end of INP concentrations.

(7) *Section 2.3.: For the multiple linear regressions, were many other models considered? What about other possibly intuitive combinations? Maybe some of the less favourable models could be included in Table 2.*

Yes, many other models have been considered following the methodological approach described in section 2.3. Still, for the sake of keeping information focused, we would rather exclude them from this paper.

(8) *Section 2.3., page 5, lines 10-12: You should consider providing more detail for the concepts in both sentences for the less statistically-minded reader.*

At the end of section 2.3 more details were added to clarify the statistical approach. The end of this paragraph now reads as: "Given an ideal model $y \sim x_1 + x_2$ (x_1 and x_2 used to derive the dependent variable y) it is possible to test whether f.i. x_2 is linearly linked to y. To do that,

the residuals of the regression of y with x_2 are plotted against the residuals of the regression of x_1 with x_2. A linear distribution of the residuals confirms the correctness of the linear relationship between x_2 and y. On the contrary, the presence of a different trend implies a different relationship between x_2 and y, like, for example, a quadratic one. Interactions between independent variables were tested as well as potential ways to improve the models. This means that the additional factor $x_1 \cdot x_2$ was inserted in a model to test whether the effect of the independent variable x_1 (or x_2) on the dependent variable y changes according to different levels of the other independent variable x_2 (or x_1). No interaction we tested resulted in a significant improvement of the models."

(9) *Section 3.1., page 6, line 12: it would make it clearer for the reader if you explicitly state why the weak correlation suggests the washout of INP is more important.*

We changed this sentence on page 6 line 12, which now reads as: "Furthermore, the ratio $NO_y \cdot CO^{-1}$ is positively correlated with wind speed and $N_{>0.5}$ (Table 1), suggesting that at high wind speed clouds at Jungfraujoch may be charged with particles taken up during recent contact of the air mass with a land surface. This suggestion could explain larger numbers of $INPs_{-8}$ in precipitation at high $NO_y \cdot CO^{-1}$ ratios (Fig. 1)".

Likely "source" and "sink" processes are further discussed in the revised version of section 3.3 (the former section 3.4).

(10) *Section 3.2.: Could the absence of one of the seasons in the validation data have any effect on the performance of second model?*

All models performed similarly in the validation step. We cannot exclude that the "season" parameter in model-2 would have given it an advantage over other models in a validation spanning all four seasons of the second year of observations. Still, it appears as unlikely since we expect that in winter the seasonal score is 1, the temperature is generally low and f_V is also low, therefore all models should perform pretty much the same. Unique in the year 2014 was the sampling in summer of precipitation from relatively warm air masses that, according to the isotopic values, had already lost considerable amount of water prior to arrival at Jungfraujoch.

(11) *Section 3.3.: I felt this section ended a little prematurely. Maybe the implications of this could be discussed. Would this imply that the 10 L^{-1} concentration requirement is too high, or is there some other factor that means the observations are below this level?*

The former section 3.3 has been removed from the manuscript according to comment 10 by Referee #2.

(12) *Section 3.4.: I am not sure what the extensive listing of various INP sources and literature adds to the argument. Consider removal or replacement with an appropriate review paper.*

We removed the literature and substituted it with this sentence in the revised version of section 3.3, including the review paper of Després et al., 2012 (added in the reference list as well): "To different degrees, surfaces on Earth, like oceans, forests, crops, soils, freshwaters, snow packs, host organisms with ice nucleating activity (Després et al., 2012) and may contribute to the airborne population of $INPs_{-8}$.".

(13) *Section 3.4., page 8, line 2: Please explain what the several multi-day sampling campaigns were. Is this the validation data?*

Every sampling campaign lasted several days and we observed that the relationship between $INPs_{-8}$ and f_V or wind speed holds not only for the whole set of data but also in several cases within the data belonging to the same sampling campaign. Section 3.4 has been changed into section 3.3 and the sentence eliminated in the revised version of the manuscript, as reported in comment 8 Referee #2.

(14) *Section 4.: The conclusions is, I feel, too brief. I came out of this feeling that I know why you did the work, but not why I should care about the result. Please add a summary at the beginning, and provide some conclusions with regards to the models. The second half of this section (lines 15-24) reads much more like it should be at the end of the introduction – either move it there or make it more relevant to the work and results presented. The only real conclusion presented is not very impressive (line 14) – the main outcomes of the work need to be better stated and some implications.*

We added a summary of the work (page 8 line 11): "Large variability in the abundance of $INPs_{-8}$ has been found in over a hundred precipitation samples collected at Jungfraujoch (3580 m a.s.l.), with values ranging from 0.21 $INPs_{-8} \cdot ml^{-1}$ to 434 $INPs_{-8} \cdot ml^{-1}$. Strikingly, with simple multiple linear regression models based on some easily measurable environmental parameters it was possible to describe and predict up to 75 % of the observed variability in $INPs_{-8}$. All these models indicate that the variability of $INPs_{-8}$ is determined by the interaction of source and sink processes, such as the potential for air masses to pick up and transport $INPs_{-8}$ and for $INPs_{-8}$ to be removed by precipitation".

The sentence starting at page 8 line 14 now reads: "Based on the results of the present study, INPs active at moderate supercooling are expected to be abundant whenever high wind speed coincides with first (initial) precipitation from an air mass.". Page 8 line 21 now reads as: "Due to this frequent co-occurrence,…"

The former section 3.4 (3.3 in the revised version) has been changed according to comment 8 Referee #2 and should provide a better bridge with the conclusions. As a generalizable conclusion, we expect large abundance of $INPs_{-8}$ at the passage of a front, which also represents the best context to concentrate sampling effort to study the impact of $INPs_{-8}$ on precipitation.

(15) *Table 2: Consider adding the actual fit parameters to the table. I'm not sure that you should include the linear-scale values, as the proportionality of these depends on where you are in the log scale – taking the log-residual of something at 10^3 and then re-linearising relative to 10^0 it does not give the actual linear value of the residual.*

All statistics for the analysis of the residuals have been done on the log-transformed values. In brackets we wanted to report the corresponding factor of error estimate on linear scale, not the median $INPs_{-8} \cdot ml^{-1}$ residual value. That is to say, if a median residual is 1 on the $\log_{10}$ scale, we can expect up to 1 order of magnitude error on a linear scale. To make the concept clearer, we modified the caption of table 2 as follows: "…The "residuals" column shows the median residual value "med" and the maximum absolute residual as maximum estimation error "ABS" (the corresponding factors of error estimate on linear scale are shown in brackets)."

(16) *All figures: please add error bars.*

This is an important suggestion, also because calculations of errors are rarely reported. In all figures we added errors bars associated to the concentrations of $INPs_{-8}$, coherently also with comment 1 and 2 Referee #2. Error bars have been cautiously calculated propagating the error associated with the method of droplet freezing (based on the error in counting frozen and unfrozen tubes, assuming a Poisson distribution) into the error associated with the temperature measurement.

To maintain the readability of figures, errors bars are shown close to the graphs for concentrations of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ and this has been indicated in the figure captions.

A description of the calculation of errors has been introduced in section 2.1: "Error bars for values of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are shown in the figures of the article. Confidence intervals were calculated as errors in counting frozen tubes, following Poisson's distribution (depending on the number of frozen tubes, they account for 30 % to 50 % increase or decrease of the calculated concentrations of $INPs_{-8}$). These intervals were propagated into the uncertainty associated with the maximum error in the determination of the freezing temperature of the tubes of ± 0.2 °C (assuming a doubling of INPs per °C of decrease in the freezing temperature, an error of 0.2 °C accounts for a change in 14 % of the measured concentrations) to provide more cautious confidence intervals".

(17) *Figure 1: For the classified data it might be helpful to add the number of data points per class somewhere. Could the number of data points in any class be high enough to cause any potential bias to the fits? Is the data from year 2 included? If so show it with a different symbol (add it if not shown). This would help with the discussion in section 3.2.*

Data from year 2 were formerly not included since the correlations presented are based only on data from year 1. Green squares have been added to represent data from year 2 to improve the discussion in section 3.2, but were not used for the correlations since they are exclusively employed to validate the data. Reference to figure 1 has been introduced in the section 3.2, while the caption of figure 1 has been adapted to the modifications and now reads: "Relationships between $INPs_{-8} \cdot ml^{-1}$ and singular environmental parameters for the first year of observations (black dots, n = 84, exception for the ratio $NO_y \cdot CO^{-1}$ with n = 51 due to missing data of trace gases). Shown are R^2 values, the coefficient of Spearman's correlation r_s and its probability p_s calculated for the data belonging to the first year of observations. f_V indicates the remaining fraction of water vapour in a precipitating air mass. In the panel "season score" 1 is for winter, 2 for spring, 3 for autumn and 4 for summer. In the panel "source region score" 1 is for Northern Europe, 2 for mixed conditions and 3 for Southern Europe. For both "season score" and "source region", values of Kruskall-Wallis' test

probability p_{kw} are shown instead of R^2. Data belonging to the second year of observations are represented as green squares (n = 15, n = 12 for the ratio $NO_y \cdot CO^{-1}$).Error bars associated to the measurement of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are represented close to the graphs for clarity."

Concerning the classified data, these are the numbers of data per class for year 1:

- Season: $n_1 = 29$; $n_2 = 25$; $n_3 = 11$; $n_4 = 19$
- Source region: $n_1 = 29$; $n_2 = 32$; $n_3 = 23$

The presence of unbalanced groups seems to be more relevant for season, but in the opposite direction as indicated. The seasons with more $INPs_{-8}$ were those with fewer measurements. To cover the point of unbalanced groups, we conservatively substituted the R^2 value with the probability associated with Kruskall-Wallis non-parametric test for comparison among groups, which considers the presence of different numbers of data in the groups. Despite this, its results do not differ much from other statistical tests. Coherently, this test has been introduced in section 2.3, page 4 line29: "For the categorical parameters "season score" and "source region score" the results of parametric regression were conservatively substituted with Kruskall-Wallis non-parametric test for the comparison among groups, because this test takes into account the presence of different numbers of data among groups."

Given the absence of a significant change in the statistical results and with the aim of keeping the figure and the figure caption as clear as possible, we would rather not indicate the number of data per group in the figure.

(18) *Figure 2/3: suggest adjusting the symbols such that the paper can be printed and understood in black and white.*

Symbols have been changed to facilitate reading in black and white. The figure has been split into 2 separate figures, to improve readability. The captions of the figures have been changed accordingly, as well as the references to them in the main text body.

(19) *Figure 3: This figure isn't really discussed in the text. I would consider either expanding its presence in the text or removing it.*

Figure removed.

Technical Comments:

(20) *Page 3, line27. Suggest insert "of" before CO, and changing "values" to concentrations, and deleting "the concentrations of" from the middle of the sentence: "...hourly concentrations of CO and total reactive...".*

Done, now the sentence reads as: "…Empa also provided hourly concentrations of CO and total reactive nitrogen NO_y in the air."

(21) *Page 4, line 14: insert a comma after "season score".*

Done.

(22) *Page 5, line 41: Citation for Wright et al is misspelt.*

Corrected the spelling.

(23) *Page 6, line 33: Delete "the dots for".*

Done.

(24) *Page 7, line 15: Please re-write the sentence so that the paragraph does not start with the word "Thus".*

Substituted "Thus, …" with "As a consequence, …".

(25) *Page 7, line 18: Delete ", also for validation".*

The whole sentence starting at page 7, line 17, has been modified into: "Model 1, based only on the two parameters f_V and wind speed, provided better results in predicting the variability of $INPs_{-8}$ observed during the second year of sampling, producing lower absolute errors, less than one log unit (Table 2)."

(26) *Page 7, line 37: "DeMott", not "De Mott" (please check elsewhere as well)*

Done and checked elsewhere.

References mentioned in the reply

Després, V. R., Huffman, J. A., Burrows, S. M., Hoose, C., Safatov, A. S., Buryak., G., Fröhlich-Nowoisky, J., Elbert, W., Andreae, M. O., Pöschl, U., and Jaenicke, R.: Primary biological aerosols particles in the atmosphere: a review, Tellus B, 64, doi.org/10.3402/tellusb.v64i0.15598, 2012.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, Atmos. Meas. Tech., 7, 129–134, doi.org/10.5194/amt-7-129-2014, 2014.

Stopelli, E., Conen, F., Morris, C. E., Herrmann, E., Bukowiecki, N., and Alewell, C.: Ice nucleation active particles are efficiently removed by precipitating clouds, Sci. Rep., 5, 16433, doi.org/10.1038/srep16433, 2015.

Referee #2:

Stopelli et al. analyze a two year time series of ice nucleating particles collected from falling precipitation at the Jungfraujoch high altitude observatory. The data are analyzed using simple correlation and multi-linear regression analysis. I read the paper with great interest. Understanding atmospheric variability of 'warm' ice nucleating particles is important and the factors that control their abundance are poorly understood. This paper presents some new insights on the subject.

However, the multi-linear regression models do not help the community to glean widely generalizable conclusions. They are location specific. Their ability to explain the temporal variation in the data does not rise beyond an order of magnitude estimate.

The conclusions are overstated. This work shows that there is some statistical correlation between INP @-8C in precipitation and temperature, fv, wind speed, source region, N500, and source region. Understanding the uncertainties in the data, the physical mechanisms driving these correlations, and the covariance matrix between the identified parameters is needed. Any claim about the role of INP in precipitation and impact on the water cycle is not supported by this work needs to be removed from the paper. The authors need to more clearly articulate the utility and limitations of the proposed statistical models. Notwithstanding a report on the inferred correlations merits publication to guide the experimental design of future studies.

Comments:

(1) *The dynamic range of the measured signal log(INP @ -8C) is 2.5 orders of magnitude, ranging from -0.5 to 2 in Fig. 2. The majority of data fluctuate between -0.5 and 0.5, and so do the residuals of the model. The statistical model is successful in identifying the high INP episodes Jun/Aug 2013 and May 2014. Other fluctuations seem to be mostly noise in either the data, the model, or both. I have several question regarding Fig. 2 and the model. What is the uncertainty in the concentration (temperature) data? Were frequent pure water tests run to determine the measurement noise?*

Blanks were periodically prepared and analysed to test the quality of our sampling and analyses. Out of 29 blanks analysed during the campaigns mentioned in the paper, only 2 of them presented some activity at -8 °C (0.11 $INPs_{-8} \cdot ml^{-1}$), indicating that only very low concentrations of INPs could be influenced by our method of analysis. More detail on the methodology and on the blank preparation has been added at the end of section 2.1: "Snow samples were analysed for the concentrations of $INPs_{-8}$ directly on site, using the automated drop freeze apparatus LINDA loaded with 52 tubes containing 100 µl of sample each (prepared adding 2 ml of 9 % NaCl sterile solution to 18 ml of sample and gently shaken, to ensure a final physiological saline concentration and improve the detection of freezing events, dilution 1:1.1, Stopelli et al. 2014; Stopelli et al., 2015). Blanks were periodically prepared distributing sterile Milli-Q water onto the rinsed tin and analysed with the same procedure as the snow samples, with 200 µl per tube to obtain more restrictive results. Out of 29 blanks analysed during the sampling campaigns reported here, only 2 blanks contained 0.11 $INPs_{-8} \cdot ml^{-1}$, confirming the accuracy of our analyses."

Further details on errors associated to measurements are provided in comment 2 Referee #2 and comment 16 Referee #1. We also think that some of the fluctuations for low values depend on the fact that the error related to counting frozen tubes is larger for low counts.

(2) *INP spectra can be steep functions of temperature. How do small errors in temperature propagate into INP @-8 °C?*

The precision of the measurements with LINDA apparatus is better than 0.2 °C (Stopelli et al., 2014). If the number of INPs active at a certain temperature doubles with each 1 °C decrease, then the errors associated to the determination of $INPs_{-8}$ range between +15 % and -13 % (corresponding to the numbers of INPs active at -8.2 and -7.8 °C respectively). Such values have been combined with the error associated to counting frozen tubes. This is described in a new paragraph is section 2.1 and errors have been added as reference close to figures of the article, as suggested in comment 16 Referee #1.

(3) *It seems that the calibration time-series for the model is at higher resolution than the data? (Trying to count green and black dots and get different numbers).*

The resolution of observed and modelled data is the same. We hope this is clearer in the revised version of the manuscript with the improved quality of the figures according to comment 18 Referee #1.

(4) *How does measurement noise, in principle, affect the multi linear regression analysis? Residuals can be interpreted as either measurement error or insufficient number or assumed physical relationship of the proposed explanatory variables. How can these be separated?*

Errors associated with measurements have been discussed in point 1 and 2 Referee #2 and point 16 Referee #1. Their relative magnitude is almost constant over several orders of magnitude, which means that errors are equally distributed on all measurements. Also residuals present no specific trend. No physical relationship has been assumed. Concerning "*insufficient number*": if it is related to the number of samples, the dataset employed seems sufficient to us; if it is related to low numbers of $INPs_{-8} \cdot ml^{-1}$, this has been treated at point 6 Referee #1. We agree that in models residuals can contain multiple factors, which cannot be easily separated.

(5) *The highest INP were observed for very high wind speeds (60-80 km* h^{-1}*). Were precautions taken that resuspended snow and wind-blown aerosol did not contaminate the measurements?*

We have several hints to exclude the fact that high wind speed massively impacted our measurements by re-suspending snow. Firstly, the tin we used to sample precipitation was placed horizontally on the terrace of the Observatory to maximize the recovery of falling snowflakes instead of those floating across the terrace with wind. Secondly, in a recent paper (Lloyd et al., 2015) it is indicated that snow blown from the surfaces surrounding the

observatory plays a role when ice particles (IPs) have small concentrations and such surfaces can be a source of secondary IPs. In our case we measure INPs by immersion freezing and not IPs or secondary IPs, moreover we observe the opposite behavior with more INPs in association with high wind speed. Thirdly, may it be that some wind-blown INPs get collected, these should have been freshly deposited on the snow surfaces around the station by the same air masses originating precipitation above the station, which makes them not much different from the one we collect on the terrace of the observatory.

(6) *Even if the source is local, how could local vs. long-range sources be distinguished?*

This is an interesting question, which cannot be entirely disentangled by measurements done at a single observatory. We tried to obtain some information on the remoteness of sources using the $NO_y \cdot CO^{-1}$ ratio and the source sensitivity plots calculated with FLEXPART model in backward mode. In the revised version of the manuscript, we indicate that under high wind speeds particles from more local sources may be easily uplifted, based on the correlation among wind speed, $N_{>0.5}$ and $NO_y \cdot CO^{-1}$ (comment 9 Referee #1). Concerning the potential impact of wind-blown ice crystals, we conceive it as negligible (please refer to the answer to comment 5 Referee #2). A precise quantification of how much a specific source of particles contributed to the $INPs_{-8}$ measured at Jungfraujoch and how far it was from the station is still a goal to be reached. Ideas on potential ways to tackle this issue in future studies are appreciated.

(7) *It is difficult to foresee all the possible cross-correlations. One of the strongest predictors for high INP seems to be temperature (even though the R^2 is low). Values of log(INP) > 1 only under the condition that temperature is at 0 °C. Thus the ability of the model to guess the high INP events is to guess when it is warm, which correlates with season, source region, RH, N>500. Assigning explanatory power to any of the variables in terms of a physical mechanism is likely erroneous.*

In the article, no precise and detailed physical mechanism is attributed to any of the variables. On the contrary, we tried to understand which variables are best correlated with $INPs_{-8}$ and, looking at correlations among variables and at the 3 best models, we tried to find possible driving forces for the variability of INPs. We propose the idea that $INPs_{-8}$ are controlled by source and sink driving forces and that the best proxies for such "black box" mechanisms are f_V and wind speed. This leads to thinking that the frontal passage coincides with the best conditions where to expect large numbers of $INPs_{-8}$. Please refer to the following point 8 Referee #2 for better explanations.

Concerning temperature, it could be a good proxy of "sink" mechanisms. Still, high-INP events do not always coincide with warm air masses. Over the second year of observation exactly the contrary has been registered. We hope that the addition of these points in figure 1 as green squares helps clarifying this (comment 17 Referee #1). The fact that around 0 °C all values of $INPs_{-8}$ are possible doesn't make of temperature an ideal descriptor of their abundance. Therefore, temperature is an important parameter but too locally constrained and does not provide any information on the precipitation history of an air mass, which seems crucial.

To comment on the threshold trend found for temperature, line 27 on page 5 in the text has been rearranged and expanded: "A better linear fit suggests that f_V is a factor capable of better

representing the temporal variability in $INPs_{-8}$ than air temperature, which shows a threshold trend. Specifically, it is possible to find more than 10 $INPs_{-8} \cdot ml^{-1}$ in precipitation for temperatures around 0 °C, indicating that when at the station the temperature is warm then also the temperature of precipitation formation in clouds above the Station can be compatible with residual large abundance of $INPs_{-8}$, but not exclusively associated only to large values of $INPs_{-8}$. Therefore, whilst air temperature appears more like a local snapshot-value for the potential activation of $INPs_{-8}$, f_V is a broader descriptor of the cumulative precipitation history of an air mass".

(8) *It is still unclear to this referee what the potential application of the statistical would be. A parameterization for a model? Improved process level understanding? Neither approach would seem promising to me without much more analysis. Perhaps the authors have another use in mind?*

Thank you for this point, which allows us to better clarify the meaning of our work. There is large variability in the numbers of INPs in precipitation, specifically for those active at moderate supercooling (Petters and Wright, 2015). With our work we want to understand which factors are responsible for such variability and point at the best conditions where and when to expect large abundance of $INPs_{-8}$.

Here we built multiple linear regression models combining parameters which a priori were already suspected to impact the abundance of $INPs_{-8}$ in precipitation based on previous observations and studies. This can help in assessing the role of such parameters in connection with $INPs_{-8}$. Results are certainly site specific, but this just means that the coefficients of the linear regression are likely to be site specific. There are important more general results which emerge from the comparison of the 3 best models:

- Wind speed is a necessary component in all 3 models and has a relation with $N_{>0.5}$ and $NO_y \cdot CO^{-1}$ (Table 1);
- Combining other parameters with wind speed, 3 models with a similar predictive power were created;
- f_V, temperature, season, source region, $N_{>0.5}$ are these further parameters and they are also well correlated among themselves (Table 1), indicating that they are all connected to the same process. It is therefore easy to link them to the processing of particles in the atmosphere. In this perspective, season and source region seem to relate to the potential of an air mass to reach Jungfraujoch with prior little precipitation;

These results point at the more generalizable fact that two driving forces are responsible for the large variability of the abundance of $INPs_{-8}$ in precipitation: the "sink" of particles, to which f_V, temperature, season, source region, $N_{>0.5}$ are related, and the "source" of particles, which seems to be linked to wind speed and partly to $N_{>0.5}$. The most synthetic and best performing model including these two driving forces is the one based on f_V and wind speed. Considering these results, we can generalize that the best condition to find large numbers of $INPs_{-8}$ are represented by the passage of a front, where meteorological conditions are also favourable to the onset of precipitation. This also indicates the best conditions where to conduct future measurements.

To clarify these concepts, several modifications have been made to the manuscript:

- page 2 line 24 the sentence now reads: "Prediction of the quantity of $INPs_{-8}$ provides useful means to understand the factors responsible for their large variability in precipitation (Petters and Wright, 2015) and to indicate the circumstances when and where $INPs_{-8}$ may

be sufficiently abundant to impact the formation of the ice phase in clouds and conduce to precipitation."

- page 7 line 33 section title changed into: "3.3 Source and sink effects"
- the whole section 3.4 (now 3.3) has been modified and now reads: "Even if the linear coefficients are site-specific, the 3 models presented in this paper point at important general indications. Wind speed is a necessary parameter to describe $INPs_{-8}$ in precipitation and is related to $N_{>0.5}$ and to a more recent land contact represented by the ratio $NO_y \cdot CO^{-1}$ (table 1). Other factors can be combined to wind speed obtaining models of comparable quality. These factors are f_V, temperature, season, source region, and $N_{>0.5}$ and are all well correlated among themselves (table 1). This means that their relation with $INPs_{-8}$ can be linked to the same process, which we suspect to be particle processing in precipitating clouds. This can act as "sink" force for $INPs_{-8}$ and is best represented by f_V. On the other hand, wind speed can strengthen the "source" of airborne particles. To different degrees, surfaces on Earth, like oceans, forests, crops, soils, freshwaters, snow packs, host organisms with ice nucleating activity (Després et al., 2012) and may contribute to the airborne population of $INPs_{-8}$. In this perspective, source region and seasonality may relate more to the likelihood for an air mass to reach Jungfraujoch with a lot of particles and little prior precipitation rather than to a different background number of airborne $INPs_{-8}$. Therefore it is possible to imagine that, independent from a more or less constant and widespread reservoir of $INPs_{-8}$, it is the combination of the energy of an air mass with the amount of precipitation generated by this air mass that determines the residual abundance of $INPs_{-8}$ in precipitation samples."
- Please refer to comment 14 Referee #1 for changes in the conclusion.

(9) *Relating INP in precipitation to INP in air, and INP in air feeding the cloud, is problematic due to a wide range of known artifacts. These include capture of INP by falling rain snow and chemical or physical modification of INP by the precipitation. None of these issues are acknowledged in the manuscript. Even if the explanatory power of the model were perfect and the intrinsic measurement error of LINDA was zero, what relevance do INP @-8 °C measured in precipitation have to clouds over Jungfraujoch?*

Please refer to the answer to comment 1 Referee #1.

(10) *The discussion in Section 3.3 should be removed since they discuss very specific cloud types, updrafts and meteorological conditions. It is entirely unclear to what extent the INP @-8 °C at Jungfraujoch have any role in precipitation without considering the larger scale meteorological forcing, including liquid water content, cloud top height, updraft velocity, the INP spectrum at warmer and colder temperatures, the efficiency of the competing warm rain process, and the cumulative ice phase history, including possible seeding by wind-blown ice crystals along the storm trajectory, including all mountains upwind. No information can be gleaned from this work about any of these processes.*

Our idea was not to infer to what extent $INPs_{-8}$ have a role on precipitation, but just to indicate when they may have been largely abundant in the atmosphere, according to available literature reporting aircraft observations. Nevertheless, based on information collected at Jungfraujoch we cannot resolve with such proposed level of detail all the processes happening in clouds. Therefore the former section 3.3 has been removed from the manuscript. Coherently, also the sentence on page 8 lines 12-14 has been removed.

References mentioned in the reply

Lloyd, G., Choularton, T. W., Bower, K. N., Gallagher, M. W., Connolly, P. J., Flynn, M., Farrington, R., Crosier, J., Schlenczek, O., Fugal, J., and Henneberger, J.: The origins of ice crystals measured in mixed-phase clouds at the high-alpine site Jungfraujoch, Atmos. Chem. Phys., 15, 12953-12969, doi:10.5194/acp-15-12953-2015, 2015.

Petters, M. D., and Wright, T. P.: Revisiting ice nucleation from precipitation samples, Geophys. Res. Lett., 42, 8758-8766, doi.org/10.1002/2015GL065733, 2015.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, Atmos. Meas. Tech., 7, 129–134, doi.org/10.5194/amt-7-129-2014, 2014.

Additional insertions and improvements

(1) The sentence on page 5 line 3 now reads as: "The normal distribution of independent and dependent variables is considered as not necessary for assessing the quality of multiple linear regression models, but it can improve the quality of the results of the model."

(2) Inserted the reference "Bowers et al., 2009" in page 2 line 9, page 5 line 41 and in the list of references.

(3) Moved the reference "Pöschl et al., 2009" to page 2 line 4.

(4) Moved the reference "Lindow et al., 1978" to page 4 line 17.

(5) Moved the reference "Huffman et al., 2013" to page 2 line 5.

(6) Added at the end of acknowledgements section: "Trace gases and PM_{10} measurements at Jungfraujoch are performed as part of the Swiss National Air Pollution Monitoring Network which is operated by Empa in collaboration with the Swiss Federal Office for the Environment."

(7) Substituted in the acknowledgement section: "by the European FP7 project BACCHUS (grant agreement no. 49603445)." with "by the European FP7 project BACCHUS (grant agreement no. 603445)."

(8) Changed part of the sentence on page 5 line 23 which now reads as: "…our current understanding of the factors that are related to the abundance of INPs…"

Predicting abundance and variability of ice nucleating particles in precipitation at the high-altitude observatory Jungfraujoch

Emiliano Stopelli[1], Franz Conen[1], Cindy E. Morris[2], Erik Herrmann[3], Stephan Henne[4], Martin Steinbacher[4], Christine Alewell[1]

[1]: Environmental Geosciences, University of Basel, 4056 Basel, CH
[2]: INRA, UR 0407 Plant Pathology Research Unit, 84143 Montfavet, FR
[3]: PSI, Laboratory of Atmospheric Chemistry, 5232 Villigen, CH
[4]: Empa, Laboratory for Air pollution/Environmental Technology, 8600 Dübendorf, CH

Correspondence to: E. Stopelli (emiliano.stopelli@unibas.ch)

Abstract

Nucleation of ice affects the properties of clouds and the formation of precipitation. Quantitative data on how ice nucleating particles (INPs) determine the distribution, occurrence and intensity of precipitation are still scarce. INPs active at -8 °C ($INPs_{-8}$) were observed for two years in precipitation samples at the High-Altitude Research Station Jungfraujoch (Switzerland) at 3580 m a.s.l. Several environmental parameters were scanned for their capability to predict the observed abundance and variability of $INPs_{-8}$. Those singularly presenting the best correlations with observed number of $INPs_{-8}$ (residual fraction of water vapour, wind speed, air temperature, number of particles with diameter larger than 0.5 µm, season and source region of particles) were implemented as potential predictor variables in statistical multiple linear regression models. These models were calibrated with 84 precipitation samples collected during the first year of observations; their predictive power was successively validated on the set of 15 precipitation samples collected during the second year. The model performing best in calibration and validation explains more than 75% of the whole variability of $INPs_{-8}$ in precipitation and indicates that a high abundance of $INPs_{-8}$ is to be expected whenever high wind speed coincides with air masses having experienced little or no precipitation prior to sampling. Such conditions occur during frontal passages, often accompanied by precipitation. Therefore, the circumstances when $INPs_{-8}$ could be sufficiently abundant to initiate the ice phase in clouds may frequently coincide with meteorological conditions favourable to the onset of precipitation events.

1 Introduction

Ice nucleating particles (INPs) play an essential role in the formation of precipitation on Earth, specifically on the continents, where most precipitation comes from ice- or mixed-phase clouds (Mülmenstädt et al., 2015). INPs catalyse the first aggregation of water molecules into ice crystals, which progressively grow larger by diffusion of surrounding water vapour and by collision with water droplets and other ice crystals until they reach a sufficient size to precipitate. In the absence of INPs spontaneous freezing would occur only at temperatures below -36 °C (Cantrell and Heymsfield, 2005; Murray et al., 2012).

Predicting abundance and variability of ice nucleating particles in precipitation at the high-altitude observatory Jungfraujoch

Emiliano Stopelli[1], Franz Conen[1], Cindy E. Morris[2], Erik Herrmann[3], Stephan Henne[4], Martin Steinbacher[4], Christine Alewell[1]

[1]: Environmental Geosciences, University of Basel, 4056 Basel, CH
[2]: INRA, UR 0407 Plant Pathology Research Unit, 84143 Montfavet, FR
[3]: PSI, Laboratory of Atmospheric Chemistry, 5232 Villigen, CH
[4]: Empa, Laboratory for Air pollution/Environmental Technology, 8600 Dübendorf, CH

Correspondence to: E. Stopelli (emiliano.stopelli@unibas.ch)

Abstract

Nucleation of ice affects the properties of clouds and the formation of precipitation. Quantitative data on how ice nucleating particles (INPs) determine the distribution, occurrence and intensity of precipitation are still scarce. INPs active at -8 °C ($INPs_{-8}$) were observed for two years in precipitation samples at the High-Altitude Research Station Jungfraujoch (Switzerland) at 3580 m a.s.l. Several environmental parameters were scanned for their capability to predict the observed abundance and variability of $INPs_{-8}$. Those singularly presenting the best correlations with observed number of $INPs_{-8}$ (residual fraction of water vapour, wind speed, air temperature, number of particles with diameter larger than 0.5 µm, season and source region of particles) were implemented as potential predictor variables in statistical multiple linear regression models. These models were calibrated with 84 precipitation samples collected during the first year of observations; their predictive power was successively validated on the set of 15 precipitation samples collected during the second year. The model performing best in calibration and validation explains more than 75% of the whole variability of $INPs_{-8}$ in precipitation and indicates that a high abundance of $INPs_{-8}$ is to be expected whenever high wind speed coincides with air masses having experienced little or no precipitation prior to sampling. Such conditions occur during frontal passages, often accompanied by precipitation. Therefore, the circumstances when $INPs_{-8}$ could be sufficiently abundant to initiate the ice phase in clouds may frequently coincide with meteorological conditions favourable to the onset of precipitation events.

1 Introduction

Ice nucleating particles (INPs) play an essential role in the formation of precipitation on Earth, specifically on the continents, where most precipitation comes from ice- or mixed-phase clouds (Mülmenstädt et al., 2015). INPs catalyse the first aggregation of water molecules into ice crystals, which progressively grow larger by diffusion of surrounding water vapour and by collision with water droplets and other ice crystals until they reach a sufficient size to precipitate. In the absence of INPs spontaneous freezing would occur only at temperatures below -36 °C (Cantrell and Heymsfield, 2005; Murray et al., 2012).

The scarcity of data about the atmospheric abundance and distribution of INPs prevents a quantitative assessment of their effect on cloud properties, on the development of precipitation and subsequently on climate. Several studies have shown the co-occurrence of INPs from local or faraway sources with precipitation at sites in the Amazon forest (Pöschl et al., 2009; Prenni et al., 2009), in the Sierra Nevada region (Creamean et al., 2013) and at a forested site in Colorado (Huffman et al., 2013; Prenni et al., 2013). The prediction of atmospheric concentrations of INPs from more easily accessible parameters would allow a more thorough evaluation of the influence of INPs on clouds and precipitation. This approach merits attention in light of results showing the correlation of specific meteorological and environmental parameters with the abundance of INPs, such as air temperature (Conen et al., 2015), wind speed (Jiang et al., 2014; Jones and Harrison, 2004), relative humidity (Bowers et al., 2009; Wright et al. 2014), season and geographical source (Christner et al., 2008), and the abundance of airborne particles of micrometre size (DeMott et al., 2010). In addition, we have recently shown that the abundance of INPs active at moderate supercooling negatively correlates with the amount of water that has been lost from an air mass prior to sampling (Stopelli et al., 2015). All these studies indicate statistical relations between INPs and the mentioned parameters, but each tends to focus predominantly on the role of a single parameter.

Here our objective is to describe and foresee the variations in the concentration of INPs active at -8 °C or warmer ($INPs_{-8}$) in falling precipitation at the high-altitude observatory Jungfraujoch (Swiss Alps, 3580 m a.s.l.) by means of multiple linear regression models. $INPs_{-8}$ are of particular interest since it has been proposed that the ice phase in clouds could be initiated by relatively few $INPs_{-8}$ (10 m^{-3} or less) through the Hallet-Mossop process of riming and ice splintering (Crawford et al., 2012; Mason, 1996). To attain our objective we firstly identified the strongest predictors for the abundance of $INPs_{-8}$ in precipitation among all the environmental parameters measured at the observatory. Secondly, we implemented these predictors in 3 multiple linear regression models built on the temporal variations in $INPs_{-8}$ occurred during the first year of observations (n = 84). The predictive power of these statistical models was subsequently tested on an independent set of samples from the second year of measurements (n = 15). Prediction of the quantity of $INPs_{-8}$ provides useful means to understand the factors responsible for their large variability in precipitation (Petters and Wright, 2015) and to indicate the circumstances when and where $INPs_{-8}$ may be sufficiently abundant to impact the formation of the ice phase in clouds and conduce to precipitation.

Deleted: guess

2 Methods

2.1 Sample collection and analysis of INPs

Falling snow was collected at the High Altitude Research Station Jungfraujoch in the Swiss Alps (7°59'06'' E, 46°32'51'' N, 3580 m a.s.l.) from December 2012 until October 2014. A total of 106 precipitation samples were collected over these two years, with a median sampling duration of about 2 hours per sample (sampling time between 1.5 and 8 hours), depending on the intensity of the precipitation events. We started sampling campaigns when the forecasts predicted 2 or more days of precipitation to assure the collection of several samples during each campaign. Samples were collected with a Teflon-coated tin carefully rinsed with ethanol and sterile Milli-Q water to avoid cross-contamination.

Deleted: The station was always inside clouds while sampling, allowing us to collect falling snow as close as possible to where it formed.

Snow samples were analysed for the concentrations of $INPs_{-8}$ directly on site, using the automated drop freeze apparatus LINDA loaded with 52 tubes containing 100 µl of sample each (prepared adding 2 ml of 9 % NaCl

sterile solution to 18 ml of sample and gently shaken, to ensure a final physiological saline concentration and improve the detection of freezing events, dilution 1:1.1, Stopelli et al. 2014; Stopelli et al., 2015). Blanks were periodically prepared distributing sterile Milli-Q water onto the rinsed tin and analysed with the same procedure as the snow samples, with 200 µl per tube to obtain more restrictive results. Out of 29 blanks analysed during the sampling campaigns reported here, only 2 blanks contained 0.11 $INPs_{-8} \cdot ml^{-1}$, confirming the accuracy of our analyses.

Error bars for values of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are shown in the figures of the article. Confidence intervals were calculated as errors in counting frozen tubes, following Poisson's distribution (depending on the number of frozen tubes, they account for 30 % to 50 % increase or decrease of the calculated concentrations of $INPs_{-8}$). These intervals were propagated into the uncertainty associated with the maximum error in the determination of the freezing temperature of the tubes of ± 0.2 °C (assuming a doubling of INPs per °C of decrease in the freezing temperature, an error of 0.2 °C accounts for a change in 14 % of the measured concentrations) to provide more cautious confidence intervals.

The collection of precipitation allows the sampling of INPs which either formed precipitating ice particles or were scavenged by precipitation. It is difficult to distinguish between these contributions in field studies, where scavenging, riming and crystal growth by vapour deposition can alter the abundance of INPs in precipitation. Nevertheless, the station was always inside clouds while sampling, allowing us to collect falling snow as close as possible to where it formed. Furthermore, precipitation was immediately analysed, in order to minimise the chance for biases due to artefacts like the production (f.i. the release in solution of INPs or cellular multiplication) and the loss (f.i. settling or increased molecular weakness of biological INPs detached from mineral and soil dust) of $INPs_{-8}$

2.2 Parameters related to the concentration of INPs

To analyse and understand more on the factors responsible for the variability of $INPs_{-8}$ in precipitation, several environmental parameters were considered in relation with the number of $INPs_{-8}$.

INPs are efficiently removed by precipitating clouds (Stopelli et al., 2015). Therefore, important information on the residual abundance of INPs in rain and snow samples is contained in the value of the residual fraction of water vapour in the sampled air mass f_V. Water molecules containing the stable isotope ^{18}O have a greater propensity to condense, hence to precipitate, than those containing the more abundant stable isotope ^{16}O. Consequently, the ^{18}O:^{16}O ratio (indicated as δ) in an air mass decreases during precipitation. f_V can be easily calculated comparing the isotopic ratio of the initial water vapour content of an air mass at the moment of its formation with the ratio at the moment of sampling according to Rayleigh's fractionation model (IAEA, 2001):

$$\frac{\delta_V}{1000} = \left(1 + \frac{\delta_{V,0}}{1000}\right) \cdot f_V^{\alpha_{L/V}-1} - 1$$

In this study δ_V is the isotopic ratio of the vapour at Jungfraujoch, calculated from the isotopic ratio of sampled snow, $\delta_{V,0}$ is the modelled isotopic ratio of the vapour in an air mass at the moment of its formation in contact with seawater and $\alpha_{L/V}$ is the fractionation factor from liquid to water along the trajectory of a cloud. Further details on the calculation of these parameters are presented in Stopelli et al., 2015.

Wind speed, air temperature and relative humidity are continuously measured at Jungfraujoch by MeteoSwiss and are stored as 10-minute averages. An optical particle counter (GrimmTM, Dust Monitor 1.108) mounted in series with a heated inlet regularly measures the total number of particles with a dry optical diameter larger than

Deleted: ,

Deleted: ¶

0.5 µm ($N_{>0.5}$) (Weingartner et al., 1999;WMO/GAW, 2003). To produce robust statistics, it was important to assign a single value of air temperature, wind speed and $N_{>0.5}$ to each snow sample. These parameters had a finer temporal resolution compared to the measurements of $INPs_{-8}$, therefore they were averaged over the time interval during which each snow sample was collected. To fill gaps due to instrument failures, missing $N_{>0.5}$ values (26 out of 106 samples) were estimated by linear regression from measured PM_{10} concentrations (R^2 = 0.40, $p < 0.001$), which are continuously determined at Jungfraujoch by Empa through beta-attenuation method (Thermo ESM Andersen FH62 IR). Due to the usually low PM_{10} concentrations at Jungfraujoch, data are aggregated to hourly averages to achieve better signal to noise ratios. In this case, the PM_{10} concentration corresponding to each snow sample was calculated averaging the hourly values including the whole duration of the collection of the sample. Empa also provided hourly concentrations of CO and total reactive nitrogen NO_y in the air. The ratio $NO_y \cdot CO^{-1}$ is a common proxy of the age of an air mass, thus it was used as indicator of planetary boundary layer influence and recent land contact of air masses sampled at the observatory (Paney Deolal et al., 2013; Griffiths et al., 2014). Due to different susceptibility to photochemical transformation in the atmosphere, $NO_y \cdot CO^{-1}$ ratios decrease during transport after being emitted from anthropogenic sources. Therefore, a larger ratio of $NO_y \cdot CO^{-1}$ is associated with a more recent contact of the air masses with land surface. Threshold values in the range 0.002 to 0.008 have been proposed to distinguish between conditions influenced by planetary boundary layer and free tropospheric air masses (Fröhlich et al., 2015; Pandey Deolal et al., 2013).

Precipitation intensity ($mm \cdot h^{-1}$) was calculated by dividing the water-equivalent volume of precipitation collected in the sampling tin by its horizontal surface and the sampling duration.

Potential regions where air masses could have picked up particles on their way to Jungfraujoch were determined by the analysis of source sensitivities simulated with FLEXPART, a Lagrangian particle dispersion model used in backward mode (Stohl et al., 2005). FLEXPART was driven with analysed meteorological fields taken from the ECMWF Integrated Forecasting Systems with a horizontal resolution of 0.2° by 0.2° over the Alpine area and 1° by 1° elsewhere (more details on the specific set-up for JFJ simulations can be found in Brunner et al., 2013). A "source region score" was assigned to each sample, combining information derived from the visual inspection of potential source regions in FLEXPART plots with the prevailing wind direction during sampling. This categorical parameter was conceived to mirror the potential differences in source quality and source strength of INP populations between northern and southern Europe. Three groups were identified: North, South and mixed/uncertain conditions. *A priori* it was hypothesised that a higher score should be given to samples from Southern Europe, assuming a larger influence of warmer air masses, enriched in larger mineral dust and organic material emissions, also linked to a more prolonged duration of agricultural activities (Kellogg and Griffin, 2006; Lindemann et al., 1982; Morris et al., 2014). Therefore *a priori* the larger value should be assigned to events from South, followed by mixed conditions and by events from Northern Europe. Several combinations of values ranging from 1 to 3 (f.i. 2-1.5-1; 3-2-1,…) were tested and the best combination of values was determined through comparisons with the numbers of log($INPs_{-8}$;ml^{-1}). It corresponds to: South = 3; mixed condition = 2, North = 1.

A similar approach was used to insert the "season score", a categorical parameter mirroring the potential effects of seasonality on the release and abundances of $INPs_{-8}$. *A priori* the highest value was assigned to samples collected in summer, assuming both a larger release of soil and organic material containing INPs, associated with the growth of vegetation, agricultural activity and warmer air masses (Jones and Harrison, 2004; Lindow et al.,

Deleted: values

Deleted: the concentrations of

Deleted: T

Deleted: subsequently

Deleted: successive

Deleted: numbers

Deleted: of

Deleted:

Deleted: and

1978; Morris et al., 2014) and a greater chance for INPs to reach the observatory before being removed by precipitation (Conen et al., 2015). In the ranking summer was followed by autumn and spring as intermediate seasons, and finally by winter. Once this *a priori* classification was established, the precise values for each class were again determined by comparing different possible combinations of numbers from 1 to 9 (f.i. 3-2-2-1; 9-6-3-1; 4-3-2-1,…) with measured values of log($INPs_{-8} \cdot ml^{-1}$). The best fit with the data was found for the combination: summer = 4; autumn = 3; spring =2; winter =1.

Deleted: comparison

2.3 Statistical analyses and modelling

Univariate statistics were carried out with PAST software version 2.17. The R software version 3.0.1 was used to build multiple linear regression models (Hammer et al., 2001; R Team, 2011).

The first step in model building consisted of a preliminary screening of the environmental parameters that had a significant relation with the variability in $INPs_{-8}$. This was done considering both the results of parametric linear regression and Spearman's non-parametric correlation test. For the categorical parameters "season score" and "source region score" the results of parametric regression were conservatively substituted with Kruskall-Wallis non-parametric test for the comparison among groups, because this test takes into account the presence of different numbers of data among groups. Normal distribution of variables is required for parametric statistics. In particular, the concentrations of $INPs_{-8}$ were approximately log-normally distributed over several orders of magnitude. Therefore, they were log_{10}-transformed to normalise their distribution. This led to the exclusion of 7 of 106 samples with no measurable activity (< 0.21 $INPs_{-8} \cdot ml^{-1}$): the arbitrary assignment of small concentrations would have resulted in a bias when projected on the log scale. Similarly, the number of particles $N_{>0.5}$, precipitation intensity and the ratio $NO_y \cdot CO^{-1}$ were log_{10}-transformed to improve the distribution of their data. Non-parametric correlation was added to draw more robust and stricter conclusions, independent from parameter distributions.

Deleted: large

Multiple linear regression models were built on the parameters presenting the best correlations with $INPs_{-8}$. Criteria to build up the models were: (a) to start from the addition of two parameters which we *a priori* suspected could be descriptors of environmental processes impacting $INPs_{-8}$ in different ways, such as proxies for their production and removal; (b) to add further parameters only if resulting in a significant gain in explained variability and improved distribution of the residuals; (c) to prefer combinations of parameters weakly correlated among themselves (Table 1), to avoid collinearity.

The normal distribution of independent and dependent variables is considered as not necessary for assessing the quality of multiple linear regression models, but it can improve the quality of the results of the model. Consequently, the variables which were log-transformed for univariate statistics were kept transformed also in multiple linear models. The quality of a multiple linear regression model is evaluated by the significance of the whole model as well as of the regression coefficients of each parameter. Particular care was taken in analysing the residuals of the models. All the models presented here fulfilled the conditions of normally distributed residuals, with an average value of zero and no significant trends. Furthermore, we assumed that the parameters could be added in linear combinations. The correctness of this assumption was verified by the method of partial regression plots of the residuals. Given an ideal model $y \sim x_1 + x_2$ (x_1 and x_2 used to derive the dependent variable y) it is possible to test whether f.i. x_2 is linearly linked to y. To do that, the residuals of the regression of y with x_2 are plotted against the residuals of the regression of x_1 with x_2. A linear distribution of the residuals confirms the correctness of the linear relationship between x_2 and y. On the contrary, the presence of a different

Deleted: not

Deleted: assessment

trend implies a different relationship between x_2 and y, like, for example, a quadratic one. Interactions between independent variables were tested as well as potential ways to improve the models. This means that the additional factor $x_1 \cdot x_2$ was inserted in a model to test whether the effect of the independent variable x_1 (or x_2) on the dependent variable y changes according to different levels of the other independent variable x_2 (or x_1). No interaction we tested resulted in a significant improvement of the models.

3 Results and discussion

3.1 Model calibration

The observations used to create the models consist of 84 snow samples with measurable concentrations of $INPs_{-8}$ collected in the Swiss Alps at 3580 m altitude between December 2012 and September 2013. Measured values of $INPs_{-8}$ ranged from the lower limit of detection ($0.21 \cdot ml^{-1}$) to a maximum of $434 \cdot ml^{-1}$. Interestingly, these values are comparable to, or even greater than those recently found in cloud water samples in Central France at 1465 m altitude (Joly et al., 2014) and well within the range of values and variability observed in precipitation samples collected all around the world (Petters and Wright, 2015).

The best correlations found at Jungfraujoch agree with our current understanding of the factors that are related to the abundance of INPs in the environment (Fig. 1, black dots). In particular, the relationships with the remaining fraction of water vapour f_V and air temperature are coherent with the observation that INPs are rapidly lost by precipitating clouds, hence are more abundant at early stages of precipitation (Stopelli et al., 2015) and that colder air masses tend to be more depleted in $INPs_{-8}$ (Conen et al., 2015). A better linear fit suggests that f_V is a factor capable of better representing the temporal variability in $INPs_{-8}$ than air temperature, which shows a threshold trend. Specifically, it is possible to find more than 10 $INPs_{-8} \cdot ml^{-1}$ in precipitation for temperatures around 0 °C, indicating that when at the station the temperature is warm then also the temperature of precipitation formation in clouds above the Station can be compatible with residual large abundance of $INPs_{-8}$, but not exclusively associated only to large values of $INPs_{-8}$. Therefore, whilst air temperature appears more like a local snapshot-value for the potential activation of $INPs_{-8}$, f_V is a broader descriptor of the cumulative precipitation history of an air mass.

Wind speed is a good proxy of the energy and turbulence associated with an air mass, promoting the transport and mixing of airborne particles (Jiang et al., 2014; Jones and Harrison, 2004). This is confirmed by the correlation between wind speed and $\log(N_{>0.5} \cdot m^{-3})$ (Table 1). Wind speed is not correlated to the direction of air masses, expressed by the source region score, indicating that the local morphology plays a minor role regarding this parameter. Coherently, the correlation between $INPs_{-8}$ and $N_{>0.5}$ suggests that the more particles $N_{>0.5}$ are present in the air, the greater is also the probability of finding a greater abundance of $INPs_{-8}$. This relationship proved significant for INPs active at -15 °C or colder (DeMott et al., 2010). Here we show its validity for INPs active at -8 °C measured in precipitation at Jungfraujoch.

$INPs_{-8}$ found in precipitation confirmed the expectations to be more abundant in summer and in air masses coming from Southern Europe. Relative humidity appears as a threshold for the abundance of $INPs_{-8}$ (Bowers et al., 2009; Wright et al., 2014), with a similar distribution of the data to the one shown by temperature. Therefore, the relationship between the relative humidity and $INPs_{-8}$ may reflect the role of particle processing in the residual abundance of $INPs_{-8}$. This process can be better represented by temperature or f_V, thus preference was given to the latter parameters in building multiple linear regression models. $INPs_{-8}$ are not correlated with the

Deleted:
Deleted: parameters
Deleted: for
Deleted: improving
Deleted: ,
Deleted: but n
Deleted:
Deleted: resulted in
Deleted: .
Deleted: can influence
Moved down [2]: A better fit suggests that f_V is a factor capable of better representing the temporal variability in $INPs_{-8}$ than air temperature.
Moved (insertion) [2]
Deleted: This can be clearly seen for temperatures around 0 °C, at which values of $INPs_{-8}$ cover several orders of magnitude.
Moved down [3]: Whilst f_V is a descriptor of the cumulative precipitation history of an air mass, air temperature appears more like a local snapshot-value for the activation of $INPs_{-8}$.
Moved (insertion) [3]
Deleted: W
Deleted: f_V is a descriptor of the cumulative precipitation history of an air mass,
Deleted: .
Deleted: t

intensity of precipitation, suggesting that different amounts of precipitation can be generated per INP. The ratio $NO_y \cdot CO^{-1}$ presents a relatively low and homogeneous range of values which are related to air masses with slightly recent contact with land surfaces (the most recent threshold value presented in literature for Jungfraujoch is 0.004, -2.4 on log scale, Fröhlich et al., 2015). Nevertheless, the sampling happened always inside precipitating clouds, which suggested the occurrence of the uplift of planetary boundary layer air to the height of the station. Therefore, it is realistic to speculate that the precipitation collected was generally originating from air masses integrating several source regions and distances before reaching the observatory. Furthermore, the ratio $NO_y \cdot CO^{-1}$ is positively correlated with wind speed and $N_{>0.5}$ (Table 1), suggesting that at high wind speed clouds at Jungfraujoch may be charged with particles taken up during recent contact of the air mass with a land surface. This suggestion could explain larger numbers of $INPs_{-8}$ in precipitation at high $NO_y \cdot CO^{-1}$ ratios (Fig. 1).

The parameters presenting the best correlations with $INPs_{-8}$ were successively added into multivariate linear regression models and the three models predicting the concentrations of $INPs_{-8}$ best are:

log (INPs$_{-8}$·ml^{-1}) =

1) 2.84 * f_V + 0.02 * *wind speed (km·h^{-1})* -1.12
2) 0.36 * *season* + 0.02 * *wind speed (km·h^{-1})*+ 0.13 * *source region*-1.39
3) 0.02 * *wind speed (km·h^{-1})*+ 0.05 * *temperature (°C)*+ 0.34 * *log($N_{>0.5}$·m^{-3})* – 1.54

They were all capable of describing about 75% of the observed variability for the calibration period (year 1, Table 2) and of reproducing observations equally well, where an apparent seasonal trend with maximum values of $INPs_{-8}$ in summer is recognisable (Fig. 2, upper panel). Yet, model 1, based on two variables only - f_V and wind speed -, performed slightly better than the other two models, which are based on three parameters. It also provided the smallest maximum absolute error (Table 2). The range of potential concentrations of $INPs_{-8}$ which can be predicted from model 1 is also the closest to observations. Inserting in the model the smallest, and largest, observed values of f_V and wind speed results in a range of calculated concentrations of 0.11 $INPs_{-8} \cdot ml^{-1}$, and 750 $INPs_{-8} \cdot ml^{-1}$. Doing the same with model 2 results in a maximum value of 250 $INPs_{-8} \cdot ml^{-1}$, and with model 3 of 400 $INPs_{-8} \cdot ml^{-1}$, underestimating the range of measured concentrations for at least one event. The observed rapid changes in the abundance of $INPs_{-8}$ may explain the slightly better performance of model 1. Differences in the concentration of $INPs_{-8}$ of more than 2 orders of magnitude were found not only on a seasonal time scale, but also within the same precipitation event over a couple of hours. The variables "season" and, to a lesser extent, "source region", "temperature" and "log($N_{>0.5} \cdot m^{-3}$)" could not always reproduce such sudden changes, as can be seen from the broader distribution of these parameters in Fig. 1.

The pattern of residuals over time is almost the same for all three models (Fig. 2, lower panel). Thus, it is unlikely to result from random noise and suggests the presence of at least one further driver of the abundance of $INPs_{-8}$ in precipitation. Given the lack of any relationship with precipitation intensity, a likely candidate is the average mass (equivalent liquid volume) of hydrometeors formed by individual INPs. For snow crystals it spans over more than an order of magnitude (Mason, 1957). INPs generating larger hydrometeors, such as those grown through riming, will be diluted in a larger volume of water and result in an overestimation of modelled numbers of $INPs_{-8} \cdot ml^{-1}$. Smaller than average ice crystals will do the opposite.

3.2 Model validation

Deleted: Furthermore, the ratio $NO_y \cdot CO^{-1}$ is weakly correlated with $INPs_{-8}$, suggesting that the effect of washout of $INPs_{-8}$ may play a larger role on their abundance than the proximity of the last contact with a land surface.

Deleted: left

Deleted:

Deleted: the dots for

Deleted: left

We validated the three models with observations from 15 precipitation samples collected between May 2014 and October 2014. The values of the environmental parameters associated with these 15 independent samples (like f_V, air temperature and so on) were directly inserted in the equations of the 3 models as derived from the calibration step and used to predict values of $INPs_{-8} \cdot ml^{-1}$.

The observed concentrations of $INPs_{-8}$ during this second period of measurements ranged from $0.21 \cdot ml^{-1}$ to a maximum of $60 \cdot ml^{-1}$. Interestingly, the samples collected in 2014 presented a completely different pattern compared to the previous year of observations (Fig. 1, green squares, and Fig. 3, upper panel). The lowest concentrations of $INPs_{-8}$ were observed during summer, whilst the highest concentrations occurred in May during a Saharan dust event and in October when a cold front from Northern Europe reached Jungfraujoch (air temperature = - 16 °C). For the sampling campaigns carried out in June, July and September the local air temperature was relatively warm (between -7 and +3 °C). Still, f_V values were low, between 0.23 and 0.47, suggesting that air masses had already lost substantial parts of their initial water vapour prior to arrival at Jungfraujoch, even if season, source region and local temperature could have been favourable for an abundant residual presence of $INPs_{-8}$.

As a consequence, models 2 and 3, which are based either on season, source region or air temperature, predicted a smaller variability of INPs than observed and overestimated the low concentrations measured in summer 2014 causing larger residual values (Fig. 3 lower panel, Table 2). Model 1, based only on the two parameters f_V and wind speed, provided better results in predicting the variability of $INPs_{-8}$ observed during the second year of sampling, producing lower absolute errors, less than one log unit (Table 2).

3.3 Source and sink effects

Even if the linear coefficients are site-specific, the 3 models presented in this paper point at important general indications. Wind speed is a necessary parameter to describe $INPs_{-8}$ in precipitation and is related to $N_{>0.5}$ and to a more recent land contact represented by the ratio $NO_y \cdot CO^{-1}$ (table 1). Other factors can be combined to wind speed obtaining models of comparable quality. These factors are f_V, temperature, season, source region, and $N_{>0.5}$ and are all well correlated among themselves (table 1). This means that their relation with $INPs_{-8}$ can be linked to the same process, which can be particle processing in precipitating clouds. This can act as "sink" force for $INPs_{-8}$ and is best represented by f_V. On the other hand, wind speed can strengthen the "source" of airborne particles. To different degrees, surfaces on Earth, like oceans, forests, crops, soils, freshwaters, snow packs, host organisms with ice nucleating activity (Després et al., 2012) and may contribute to the airborne population of $INPs_{-8}$. In this perspective, source region and seasonality may relate more to the likelihood for an air mass to reach Jungfraujoch with a lot of particles and little prior precipitation rather than to a different background number of airborne $INPs_{-8}$. Therefore it is possible to imagine that, independent from a more or less constant and widespread reservoir of $INPs_{-8}$, it is the combination of the energy of an air mass with the amount of precipitation generated by this air mass that determines the residual abundance of $INPs_{-8}$ in precipitation samples.

4 Conclusions

Deleted: 2
Deleted: right
Deleted: Thus
Deleted: 2
Deleted: right
Deleted:
Deleted: the best
Deleted: describing
Deleted: , also for
Deleted: validation
Deleted: . It
Deleted: ed
Deleted: the lowest
Deleted: and its results more closely matched the observed dynamics and approached the optimal 1:1 agreement between predicted and measured values over the full range of observed concentrations of $INPs_{-8}$ (Fig. 3).
Deleted: ¶
Deleted: **3.3 Fraction of clouds with enough $INPs_{-8}$ to be conducive to the ice phase**¶ A concentration of 10 $INPs_{-8} \cdot m^{-3}$ of air has been observed through in-cloud measurement as a sufficient number to initiate the ice phase (Crawford et al., 2012; Mason, 1996). This allows calculating a conservative estimate of the fraction of observations where $INPs_{-8}$ could theoretically still have initiated the ice phase in clouds, if it had not already started earlier (note: we sampled already precipitating clouds). Considering the observed median value of 0.25 ml of precipitation from 1 m^3 of air (Stopelli et al., 2015), around 40 $INPs_{-8} \cdot ml^{-1}$ should be sufficient to indicate that a precipitation sample originated from a cloud where $INPs_{-8}$ would still have been numerous enough to induce the ice phase. Referring to the full set of samples used for calibration and validation, such a concentration was found or exceeded in 12 (10 %) of 106 samples.¶
Deleted: 4
Deleted: Open question: s
Deleted: It remains an open question whether the seasonal change in the sourc…
Moved (insertion) [1]
Deleted: the roles of
Deleted: reflect the potential
Deleted: a different
Deleted: In addition to that, the good correlations between $INPs_{-8}$ and f_V and …
Deleted: together
Deleted:
Moved up [1]: In this perspective, the roles of source region and seasonality ma…
Deleted: ¶

Large variability in the abundance of $INPs_{-8}$ has been found in over a hundred precipitation samples collected at Jungfraujoch (3580 m a.s.l.), with values ranging from 0.21 $INPs_{-8} \cdot ml^{-1}$ to 434 $INPs_{-8} \cdot ml^{-1}$. Strikingly, with simple multiple linear regression models based on some easily measurable environmental parameters it was possible to describe and predict up to 75 % of the observed variability in $INPs_{-8}$. All these models indicate that the variability of $INPs_{-8}$ is determined by the interaction of "source" and "sink" processes, such as the potential for air masses to pick up and transport $INPs_{-8}$ from several sources and for $INPs_{-8}$ to be removed by precipitation. Our investigation indicates that a large abundance of $INPs_{-8}$ in precipitation at Jungfraujoch is present whenever there is a coincidence of high wind speed and moist air mass with little or no prior precipitation. Based on the results of the present study, INPs active at moderate supercooling are expected to be abundant whenever high wind speed coincides with first (initial) precipitation from an air mass. These conditions can be met when an air mass is suddenly forced to rise, for instance at the boundary of a front or due to thermal updrafts or when crossing a mountain ridge. Specifically during the passage of a cold front, gusty winds promote the uptake of particles and the first clouds that form will still retain a large fraction of the initial water vapour of the warm air mass (Gayet et al., 2009; Wright et al., 2014). Simultaneously, physical conditions along a cold front are favourable for cloud formation. Therefore, frequent systematic coincidences of high numbers of INPs with meteorological conditions conducive to precipitation may be expected. Due to this frequent co-occurrence, the potential impact on precipitation by INPs active at slight supercooling - such as INPs of biological origin - may be larger than previously estimated. Their role in the water cycle might therefore best be studied under such conditions.

Deleted: Furthermore, we have estimated that for 10 % of our precipitation samples there would have been enough $INPs_{-8}$ to initiate the ice phase in clouds. Based on the results of the present study, INPs active at moderate supercooling are expected to play a role whenever conditions of high wind speed and first precipitation from air masses prevail

Author contributions

E.S. and F.C. did the field measurements at Jungfraujoch on the concentrations of INPs, analysed data, did statistical modelling and wrote the manuscript. E.H., S.H. and M.S. respectively provided data and support for the interpretation of $N_{>0.5}$, FLEXPART modelling, PM_{10} and trace gases and contributed to writing the paper. C.M. and C.A. provided strong conceptual frameworks and contributed to writing the paper.

Acknowledgements

We thank the International Foundation for High Alpine Research Station Jungfraujoch and Gornergrat (HFSJG) for making it possible for us to conduct our measurements at Jungfraujoch. Urs and Maria Otz, Martin and Joan Fischer provided helpful support during field activity. Corinne Baudinot measured the abundance of INPs in snow samples during the second year of observations. Dr Thomas Kuhn and Mark Rollog analysed the stable isotope ratio in our snow water samples. We thank MeteoSwiss for providing data on meteorology at Jungfraujoch. The work described here was supported by the Swiss National Science Foundation (SNF) through grant no 200021_140228 and 200020_159194. Measurements of total solid particles were performed by Paul Scherrer Institute in the framework of the Global Atmospheric Watch (GAW) programme funded by MeteoSwiss with further support provided by the European FP7 project BACCHUS (grant agreement no. 603445). Trace gases and PM_{10} measurements at Jungfraujoch are performed as part of the Swiss National Air Pollution Monitoring Network which is operated by Empa in collaboration with the Swiss Federal Office for the Environment.

The authors declare no competing financial interests.

Deleted: 49

References

Bowers, R. M., Lauber, C. L., Wiedinmyer, C., Hamady, M., Hallar, A. G., Fall, R., Knight, R., and Fierer, N.: Characterization of airborne microbial communities at a high-elevation site and their potential to act as atmospheric ice nuclei, Appl. Environ. Microbiol., 75, 5121–5130, doi.org/10.1128/AEM.00447-09, 2009.

Brunner, D., Henne, S., Keller, C. A., Vollmer, M. K., Reimann, S., and Buchmann, B.: Estimating European Halocarbon Emissions Using Lagrangian Backward Transport Modeling and in Situ Measurements at the Jungfraujoch High-Alpine Site, Lagrangian Modeling of the Atmosphere (eds J. Lin, D. Brunner, C. Gerbig, A. Stohl, A. Luhar and P. Webley), American Geophysical Union, Washington DC, 207-221, doi.org/10.1029/2012GM001258, 2013.

Cantrell, W. and Heymsfield, A.: Production of Ice in Tropospheric Clouds: A Review, B. Am. Meteorol. Soc., 86, 795–807, doi.org/10.1175/BAMS-86-6-795, 2005.

Christner, B. C., Cai, R., Morris, C. E., McCarter, K. S., Foreman, C. M., Skidmore, M. L., Montross, S. N., and Sands, D. C.: Geographic, seasonal, and precipitation chemistry influence on the abundance and activity of biological ice nucleators in rain and snow, P. Natl. Acad. Sci. USA, 105, 18854–18859, doi.org/10.1073/pnas.0809816105, 2008.

Conen, F., Rodriguez, S., Hüglin, C., Henne, S., Herrmann, E., Bukowiecki, N., and Alewell, C.: Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland, Tellus B, 67, 1–10, doi.org/10.3402/tellusb.v67.25014, 2015.

Crawford, I., Bower, K. N., Choularton, T. W., Dearden, C., Crosier, J., Westbrook, C., Capes, G., Coe, H., Connolly, P. J., Dorsey, J. R., Gallagher, M. W., Williams, P., Trembath, J., Cui, Z., and Blyth, A.: Ice formation and development in aged, wintertime cumulus over the UK: observations and modelling, Atmos. Chem. Phys., 12, 4963–4985, doi.org/10.5194/acp-12-4963-2012, 2012.

Creamean, J. M., Suski, K. J., Rosenfeld, D., Cazorla, A., DeMott, P. J., Sullivan, R. C., White, A. B., Ralph, F. M., Minnis, P., Comstock, J. M., Tomlinson, J. M., and Prather, K. A.: Dust and biological aerosols from the Sahara and Asia influence precipitation in the western U.S., Science, 339, 1572–1578, doi.org/10.1126/science.1227279, 2013.

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, P. Natl. Acad. Sci. USA, 107, 11217–11222, doi.org/10.1073/pnas.0910818107, 2010.

Després, V. R., Huffman, J. A., Burrows, S. M., Hoose, C., Safatov, A. S., Buryak., G., Fröhlich-Nowoisky, J., Elbert, W., Andreae, M. O., Pöschl, U., and Jaenicke, R.: Primary biological aerosols particles in the atmosphere: a review, Tellus B, 64, doi.org/10.3402/tellusb.v64i0.15598, 2012.

Fröhlich, R., Cubison, M. J., Slowik, J. G., Bukowiecki, N., Canonaco, F., Croteau, P. L., Gysel, M., Henne, S., Herrmann, E., Jayne, T. J., Steinbacher, M., Worsnop, D. R., Baltensperger, U., and Prévôt, A. S. H.: Fourteen months of on-line measurements of the non-refractory submicron aerosol at the Jungfraujoch (3580m a.s.l.) - chemical composition, origins and organic aerosol sources, Atmos. Chem. Phys., 15, 11373-11398, doi.org/10.5194/acp-15-11373-2015, 2015.

Gayet, J. F., Treffeisen, R., Helbig, A., Bareiss, J., Matsuki, A., Herber, A., and Schwarzenboeck, A.: On the onset of the ice phase in boundary layer Arctic clouds, J. Geophys. Res., 114, 1–15, doi.org/10.1029/2008JD011348, 2009.

Deleted: Conen, F., Morris, C. E., Leifeld, J., Yakutin, M. V., and Alewell, C.: Biological residues define the ice nucleation properties of soil dust, Atmos. Chem. Phys., 11, 9643–9648, doi.org/10.5194/acp-11-9643-2011, 2011.¶

Deleted: DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Martinez, M. D., Venero, I., Santos-Figueroa, G., Dale Stokes, M., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F., and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles. P. Natl. Acad. Sci. USA Early Edition, doi.org/10.1073/pnas.1514034112, 2015.¶

Deleted: Fröhlich-Nowoisky, J., Hill, T. C. J., Pummer, B. G., Yordanova, P., Franc, G. D., and Pöschl, U.: Ice nucleation activity in the widespread soil fungus *Mortierella alpina*, Biogeosciences, 12, 1057–1071, doi.org/10.5194/bg-12-1057-2015, 2015.¶

Griffiths, A. D., Conen, F., Weingartner, E., Zimmermann, L., Chambers, S. D., Williams, A. G., and Steinbacher, M.: Surface-to-mountaintop transport characterised by radon observations at the Jungfraujoch, Atmos. Chem. Phys., 14, 12763-12779, doi.org/10.5194/acp-14-12763-2014, 2014.

Hammer, Ø., Harper, D. A., and Ryan, P. D.: PAST: PAleontologi- cal STatistics software package for education and data analysis, Palaeontol. Electron., 4, available at: http://www.uv.es/pe/2001_ 1/past/past.pdf (last access: 06.01.2016), 2001.

Huffman, J. A., Prenni, A. J., DeMott, P. J., Pöhlker, C., Mason, R. H., Robinson, N. H., Fröhlich-Nowoisky, J., Tobo, Y., Després, V. R., Garcia, E., Gochis, D. J., Harris, E., Müller-Germann, I., Ruzene, C., Schmer, B., Sinha, B., Day, D. A., Andreae, M. O., Jimenez, J. L., Gallagher, M., Kreidenweis, S. M., Bertram, A. K., and Pöschl, U.: High concentrations of biological aerosol particles and ice nuclei during and after rain, Atmos. Chem. Phys., 13, 6151–6164, doi.org/10.5194/acp-13-6151-2013, 2013.

IAEA International Atomic Energy Agency: Environmental isotopes in the hydrological cycle. Principles and applications. Vol. 2. Atmospheric Water, available at: http://www-naweb.iaea.org/napc/ih/IHS_resources_publication_hydroCycle_en.html (last access on 06.01.2016), 2001.

Jiang, H., Yin, Y., Yang, L., Yang, S., Su, H., and Chen, K.: The Characteristics of Atmospheric Ice Nuclei Measured at Different Altitudes in the Huangshan Mountains in the Southwest China, Adv. Atmos. Sci., 31, 396-406, doi.org/10.1007/s00376-013-3048-5, 2014.

Joly, M., Amato, P., Deguillaume, L., Monier, M., Hoose, C., and Delort, A. M.: Quantification of ice nuclei active at near 0 °C temperatures in low-altitude clouds at the Puy de Dôme atmospheric station, Atmos. Chem. Phys., 14, 8185–8195, doi.org/10.5194/acp-14-8185-2014, 2014.

Jones, A. M. and Harrison, R. M.: The effects of meteorological factors on atmospheric bioaerosol concentrations-a review, Sci. Total Environ., 326, 151–180, doi.org/10.1016/j.scitotenv.2003.11.021, 2004.

Kellogg, C. A. and Griffin, D. W.: Aerobiology and the global transport of desert dust, Trends Ecol. Evol., 21, 638–644, doi.org/10.1016/j.tree.2006.07.004, 2006.

Lindemann, J., Constantinidou, H. A., Barchet, W. R., and Upper, C. D.: Plants as sources of airborne bacteria, including ice nucleation-active bacteria, Appl. Environ. Microb., 44, 1059–1063, 1982.

Lindow, S. E., Arny, D. C., and Upper, C. D.: Distribution of ice nucleation-active bacteria on plants in nature, Appl. Environ. Microb., 36, 831-838, 1978.

Mason, B. J.: The Physics of Clouds, Oxford University Press, 1957.

Mason, B. J.: The rapid glaciation of slightly supercooled cumulus clouds, Q. J. R. Meteorol. Soc., 122, 357-365, doi.org/10.1002/qj.49712253003, 1996.

Morris, C. E., Conen, F., Huffman, J. A., Phillips, V., Pöschl, U., and Sands, D. C.: Bioprecipitation: a feedback cycle linking earth history, ecosystem dynamics and land use through biological ice nucleators in the atmosphere, Glob. Change Biol., 20, 341–351, doi.org/10.1111/gcb.12447, 2014.

Mülmenstädt, J., Sourdeval, O., Delanoë, J., and Quaas, J.: Frequency of occurrence of rain from liquid- , mixed- , and ice-phase clouds derived from A-Train satellite retrievals, Geophys. Res. Lett., 42, 6502–6509, doi.org/10.1002/2015GL064604, 2015.

Murray, B. J., O'Sullivan, D., Atkinson, J. D., and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519–6554, doi.org/10.1039/c2cs35200a, 2012.

Deleted:

Deleted: Moffett, B. F., Getti, G., Henderson-Begg, K., and Hill, T. C. J.: Ubiquity of ice nucleation in lichen – possible atmospheric implications, Lindbergia, 38, 39-43, 2015.¶ Monteil, C. L., Guilbaud, C., Glaux, C., Lafolie, F., Soubeyrand, S., and Morris, C. E.: Emigration of the plant pathogen *Pseudomonas syringae* from leaf litter contributes to its population dynamics in alpine snowpack, Environ. Microbiol., 14, 2099–2112, doi.org/10.1111/j.1462-2920.2011.02680.x, 2012. ¶ Morris, C. E., Sands, D. C., Vanneste, J. L., Montarry, J., Oakley, B., Guilbaud, C., and Glaux, C.: Inferring the Evolutionary History of the Plant Pathogen *Pseudomonas syringae* from Its Biogeography in Headwaters of Rivers in North America, Europe, and New Zealand, mBio, 1, e00107-10, doi.org/10.1128/mBio.00107-10, 2010.¶

Deleted: ¶ O'Sullivan, D., Murray, B. J., Ross, J. F., Whale, T. F., Price, H. C., Atkinson, J. D., Umo, N. S., and Webb, M. E.: The relevance of nanoscale biological fragments for ice nucleation in clouds, Sci. Rep., 5, 8082, doi.org/10.1038/srep08082, 2015.

Pandey Deolal, S., Staehelin, J., Brunner, D., Cui, J., Steinbacher, M., Zellweger, C., Henne, S., and Vollmer, M. K.: Transport of PAN and NO_y from different source regions to the Swiss high alpine site Jungfraujoch, Atmos. Environ., 64, 103-115, doi.org/10.1016/j.atmosenv.2012.08.021, 2013.

Petters, M. D., and Wright, T. P.: Revisiting ice nucleation from precipitation samples, Geophys. Res. Lett., 42, 8758-8766, doi.org/10.1002/2015GL065733, 2015.

Pöschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S. R., Artaxo, P., and Andreae, M. O.: Rainforest Aerosols as Biogenic Nuclei of Clouds and Precipitation in the Amazon, Science, 329, 1513–1516, doi.org/10.1126/science.1191056, 2009.

Prenni, A. J., Petters, M. D., Kreidenweis, S. M., Heald, C. L., Martin, S. T., Artaxo, P., Garland, R. M., Wollny, A. G., and Pöschl, U.: Relative roles of biogenic emissions and Saharan dust as ice nuclei in the Amazon basin, Nature Geosci., 2, 402–405, doi.org/10.1038/ngeo517, 2009.

Prenni, A. J., Tobo, Y., Garcia, E., DeMott, P. J., Huffman, J. A., McCluskey, C. S., Kreidenweis, S. M., Prenni, J. E., Pöhlker, C., and Pöschl, U.: The impact of rain on ice nuclei populations at a forested site in Colorado, Geophys. Res. Lett., 40, 227–231, doi.org/10.1029/2012GL053953, 2013.

R Core Team: R: A Language and Environment for Statistical Computing, R Foundation for Statistical Computing, Vienna, Austria, available at: http://www.R-project.org (last access: 06.01.2016), 2011.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: the Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, doi.org/10.5194/acp-5-2461-2005, 2005.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, Atmos. Meas. Tech., 7, 129–134, doi.org/10.5194/amt-7-129-2014, 2014.

Stopelli, E., Conen, F., Morris, C. E., Herrmann, E., Bukowiecki, N., and Alewell, C.: Ice nucleation active particles are efficiently removed by precipitating clouds, Sci. Rep., 5, 16433, doi.org/10.1038/srep16433, 2015.

Weingartner, E., Nyeki, S., and Baltenspergcr, U.: Seasonal and diurnal variation of aerosol size distributions (10 < D < 750 nm) at a high-alpine site (Jungfraujoch 3580 m asl), J. Geophys. Res., 104, 809–826, doi.org/10.1029/1999JD900170, 1999.

WMO/GAW World Meteorological Organisation/ Global Atmospheric Watch: Aerosol Measurements Procedures. Guidelines and Recommendations, available at: https://www.wmo.int/pages/prog/gcos/documents/gruanmanuals/GAW/gaw143.pdf (last access: 06.01.2016), 2003.

Wright, T. P., Hader, J. D., McMeeking, G. R., and Petters, M. D.: High Relative Humidity as a Trigger for Widespread Release of Ice Nuclei, Aerosol Sci. Tech., 48, i–v, doi.org/10.1080/02786826.2014.968244, 2014.

Deleted: Pummer, B. G., Budke, C., Augustin-Bauditz, S., Niedermeier, D., Felgitsch, L., Kampf, C. J., Huber, R. G., Liedl, K. R., Loerting, T., Moschen, T., Schauperl, M., Tollinger, M., Morris, C. E., Wex, H., Grothe, H., Pöschl, U., Koop, T., and Fröhlich-Nowoisky, J.: Ice nucleation by water-soluble macromolecules, Atmos. Chem. Phys., 15, 4077–4091, doi.org/10.5194/acp-15-4077-2015, 2015.¶

Deleted: Schnell, R. C. and Vali, G.: Atmospheric ice nuclei from decomposing vegetation, Nature, 236, 163-165, 1972.¶
Schnell, R. C. and Vali, G.: World-wide source of leaf-derived freezing nuclei, Nature, 246, 212-213, 1973.¶
Schnell, R. C. and Vali, G.: Freezing nuclei in marine waters, Tellus, 27, 321–323, 1975.¶

Deleted: ¶
Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. A., Nájera, J. J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Vergara Temprado, J., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. A., and Murray, B. J.: A marine biogenic source of atmospheric ice nucleating particles, Nature, 525, 234-238, doi.org/10.1038/nature14986, 2015.

Table 1: Pairwise correlations among predictors for the concentrations of $INPs_{-8}$. The upper panel indicates the pairwise Spearman's r_s correlation coefficient, the lower panel its statistical significance (*,**,*** stand for a probability smaller than 0.05, 0.01 and 0.001 respectively).

	f_V	wind speed	temperature	$\log(N_{>0.5} \cdot m^{-3})$	season	source region	relative humidity	$\log(mm \cdot h^{-1})$	$\log(NO_y \cdot CO^{-1})$
f_V		0.10	0.88	0.33	0.89	0.42	0.72	0.07	0.13
wind speed			0.02	0.21	-0.04	0.17	0.04	0.08	0.31
temperature	***			0.18	0.85	0.58	0.70	-0.03	-0.28
$\log(N_{>0.5} \cdot m^{-3})$	**	*	*		0.35	0.32	0.28	-0.11	0.37
season	***		***	***		0.38	0.77	-0.04	0.15
source region	***		***	**	***		0.42	-0.12	-0.23
relative humidity	***		***	**	***	***		-0.12	0.23
$\log(mm \cdot h^{-1})$									0.24
$\log(NO_y \cdot CO^{-1})$		*	*	**		*	*	*	

Table 2: Summary of the main statistics employed to evaluate the quality of the models. "$\mathbf{R^2_{adj}}$" is the fraction of the observed variance reproduced by a model. It is adjusted to account for the number of variables and samples considered. All models, parameters, and constants are highly significant ($p < 0.001$), except the parameter "source region" ($p = 0.06$). "**β***" is the value of the standardised regression coefficients, expressing the relative importance of each parameter in a model. The "residuals" column shows the median residual value "med" and the maximum absolute residual as maximum estimation error "ABS" (the corresponding factors of error estimate on linear scale are shown in brackets). "MSE" is the value of the mean squared error.

	Calibration			Validation
	$\mathbf{R^2_{adj}}$	**β***	residuals	residuals
1	**0.76**	f_V: **0.62** wind speed: **0.47**	Med: -0.04 (0.9) ABS: 1.02 (10.4) MSE: 0.16	Med: -0.08 (0.8) ABS: 0.87 (7.4) MSE: 0.25
2	**0.73**	season: **0.52** wind speed: **0.52** source region: **0.12**	Med: 0.00 (1.0) ABS: 1.45 (28.4) MSE: 0.17	Med: -0.25 (0.6) ABS: 1.42 (26.1) MSE: 0.56
3	**0.74**	wind speed:**0.49** temperature: **0.48** log ($N_{>0.5} \cdot m^{-3}$): **0.26**	Med: -0.02 (0.9) ABS: 1.22 (16.5) MSE: 0.17	Med: -0.33 (0.5) ABS: 1.13 (13.5) MSE:0.43

Deleted: values

Deleted: o

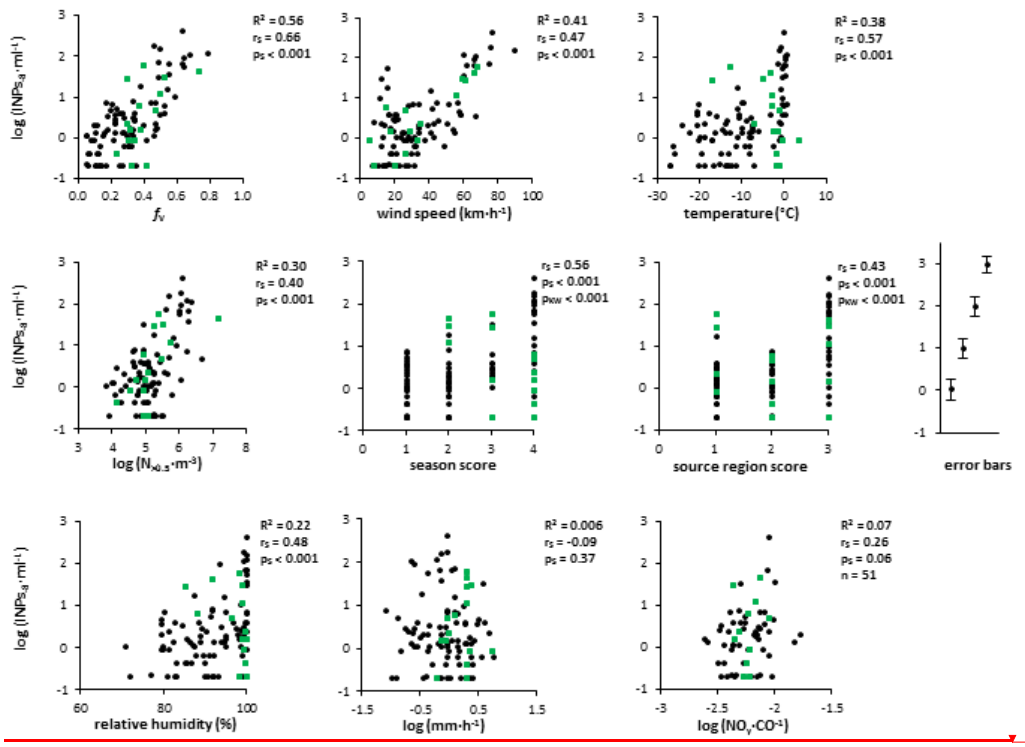
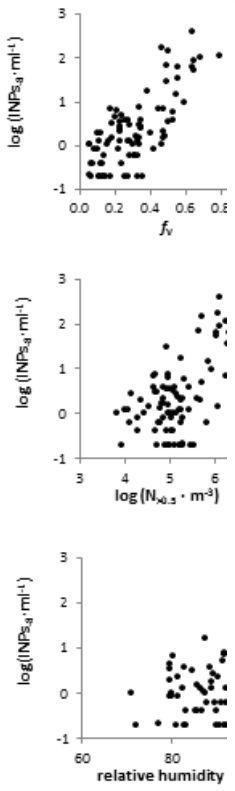


Fig. 1: Relationships between $INPs_{-8} \cdot ml^{-1}$ and singular environmental parameters for the first year of observations (black dots, n = 84, exception for the ratio $NO_y \cdot CO^{-1}$ with n = 51 due to missing data of trace gases). Shown are R^2 values, the coefficient of Spearman's correlation r_s and its probability p_s calculated for the data belonging to the first year of observations. f_V indicates the remaining fraction of water vapour in a precipitating air mass. In the panel "season score" 1 is for winter, 2 for spring, 3 for autumn and 4 for summer. In the panel "source region score" 1 is for Northern Europe, 2 for mixed conditions and 3 for Southern Europe. For both "season score" and "source region", values of Kruskall-Wallis' test probability p_{kw} are shown instead of R^2. Data belonging to the second year of observations are represented as green squares (n = 15, n = 12 for the ratio $NO_y \cdot CO^{-1}$). Error bars associated to the measurement of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are represented close to the graphs.



Deleted:

Deleted: .

Deleted: r_s,

Deleted: ,

Deleted:

Deleted: . The number of samples (n) was always 84, exception for the ratio $NO_y \cdot CO^{-1}$ (n = 51) due to missing trace gases.

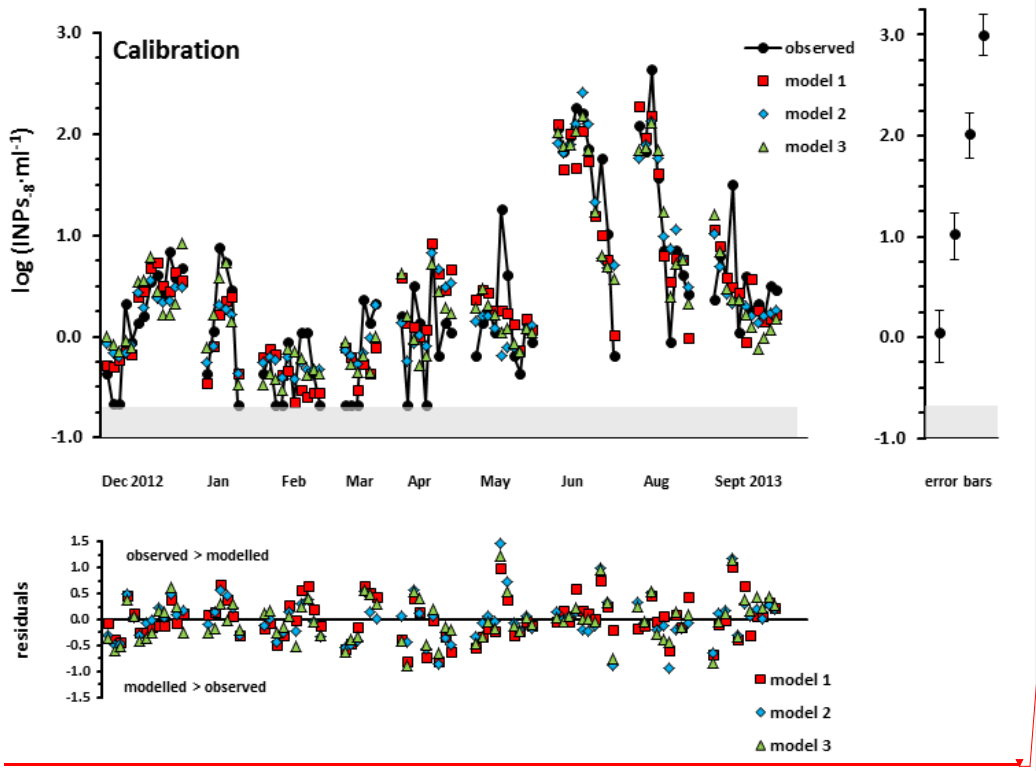
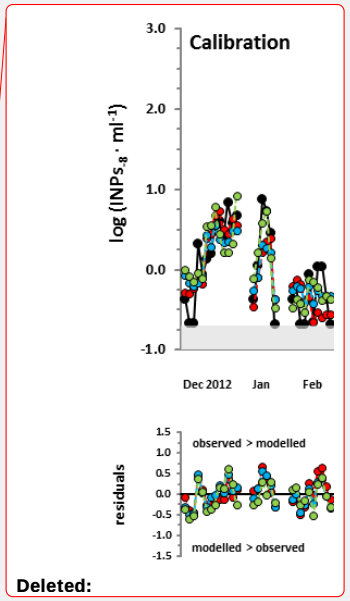


Fig. 2: Comparison of observed concentrations of $INPs_{-8}$ with the values predicted by models (absolute values in upper panels, residuals in lower panels) for the dataset used for calibration (n = 84). The grey area in the upper panel indicates the range below the detection limit of our observation method. Time proceeds from left to right, intervals are not to scale, dots belonging to the same sampling campaign are connected by lines. Error bars associated to the measurement of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are represented close to the graph.

Deleted:



Deleted: . On the left side the results of model calibration are presented, on the right side those for the validation of the models.

Deleted: s

Deleted: dashed

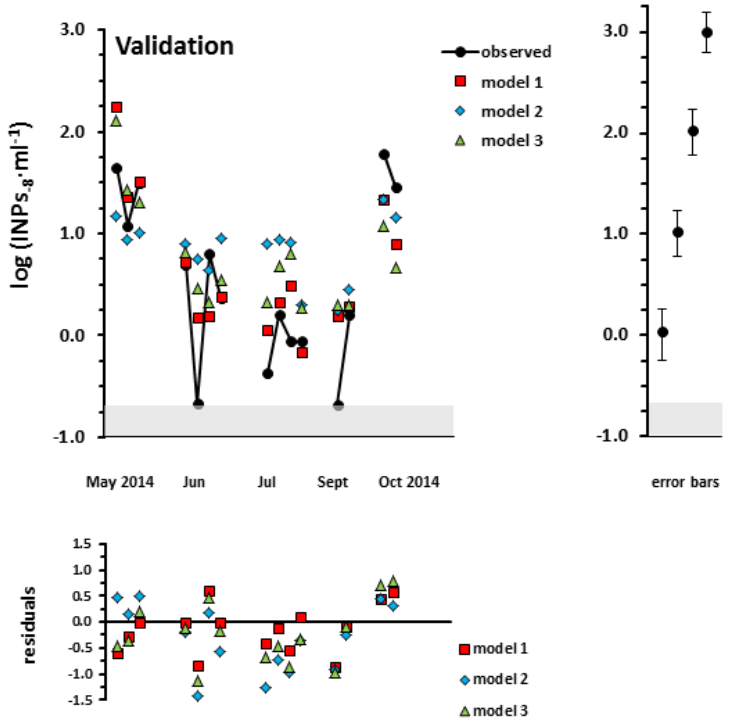


Fig. 3: Comparison of observed concentrations of $INPs_{-8}$ with the values predicted by models (absolute values in upper panels, residuals in lower panels) for the dataset used for validation (n = 15). The grey area in the upper panel indicates the range below the detection limit of our observation method. Time proceeds from left to right, intervals are not to scale, dots belonging to the same sampling campaign are connected by lines. Error bars associated to the measurement of 1, 10, 100, 1000 $INPs_{-8} \cdot ml^{-1}$ are represented close to the graph.

Deleted: ¶

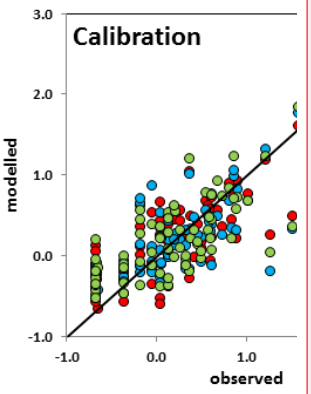


Fig. 3: Fitting of modelled concentrations of $INPs_{-8}$ to the observed values, expressed on the log scale. The black line represents the perfect 1:1 correspondence. Specifically for the validation, it is possible to observe how model 2 and 3 tend to overestimate the lowest concentrations of $INPs_{-8}$ measured during summer 2014.